Neutron scattering

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Outline

Why is neutron scattering important?

Peculiarities of neutrons

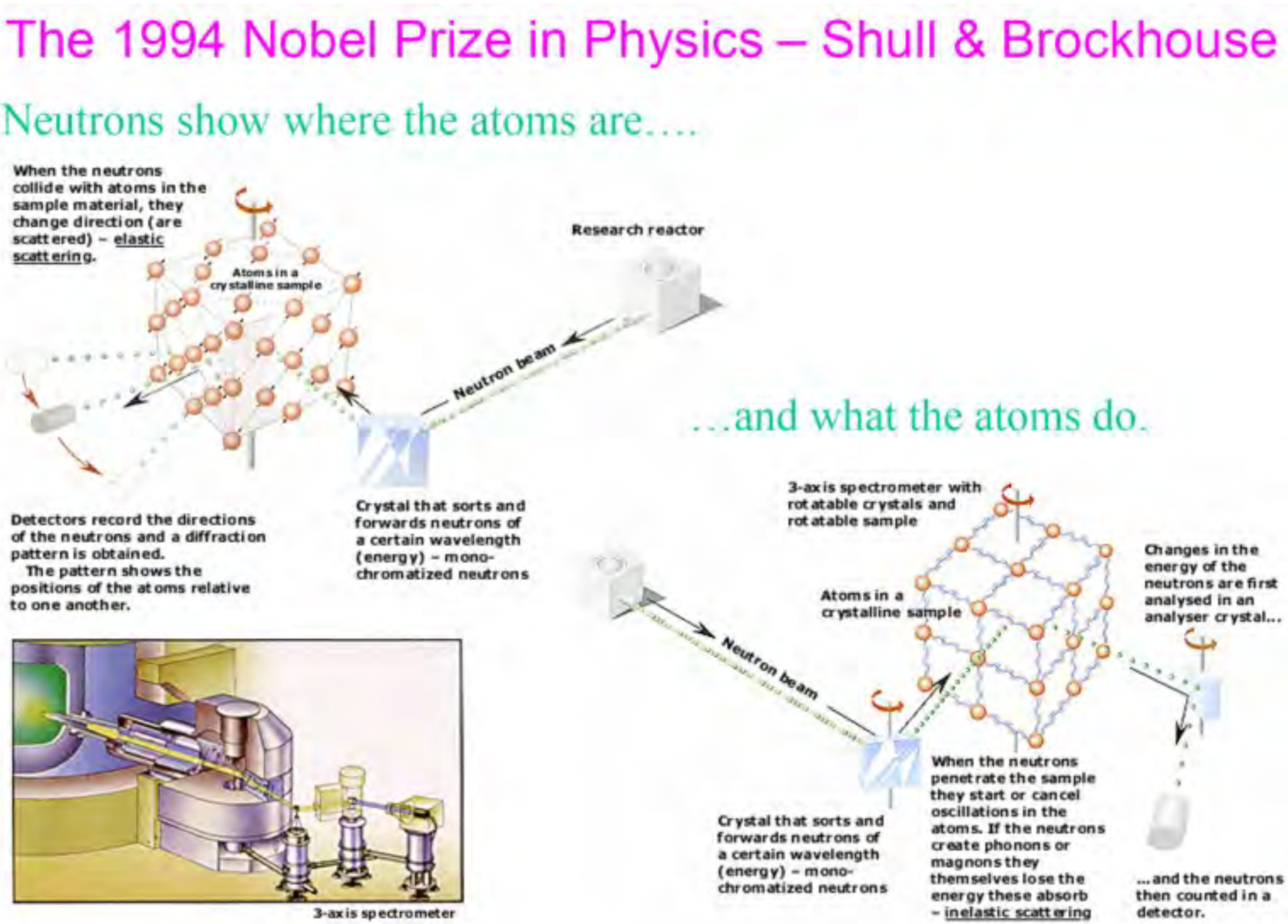
How does it work?

What do we measure?
How do we measure?

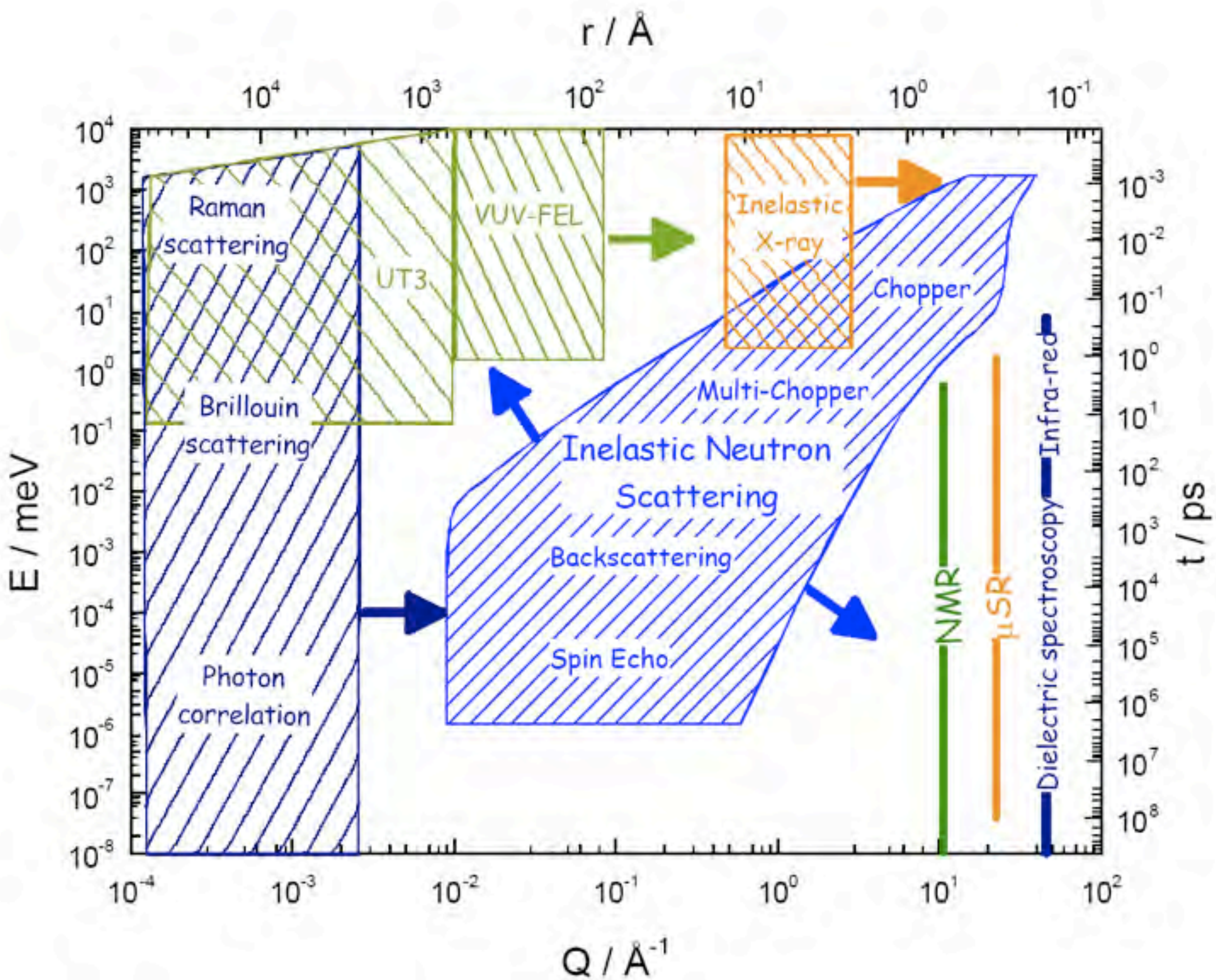
What is it good for?

Applications

Neutron scattering



Time and length scales



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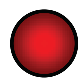
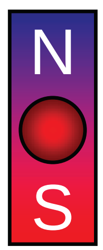
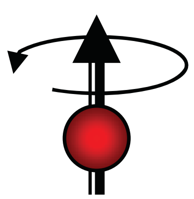
Why neutrons?



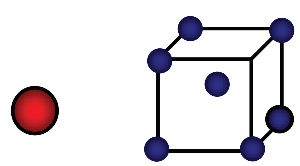
Neutrons are neutral: high penetation power, non-destructive



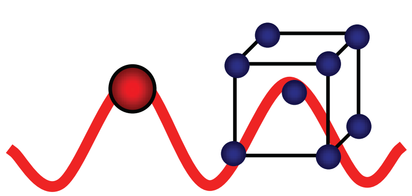
Neutrons have a magnetic moment: magnetic structures and excitations



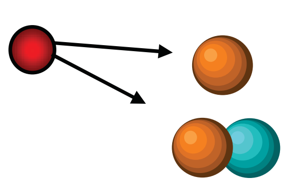
Neutrons have a spin: polarized neutrons, coherent and incoherent scattering, nuclear magnetism



Neutrons have thermal energies: excitation of elemenatry modes, phonons, magnons, librons, rotons, tunneling, etc.



Neutrons have wavelengths similar to atomic spacings: structural information, short and long range order, pore and grain sizes, cavities, etc.



Neutrons see nuclei: sensitive to light atoms, exploiting isotopic substitution, contrast variation with isotopes

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The neutron

Properties:

Life time:	$T_{1/2}$= 890 s
Mass:	m = 1.675 * 10 $^{-27}$ kg
Charge:	Q = 0
Spin:	S = $\hbar/2$
Mag. Moment:	μ/μ_n = -1.913

Dispersion:

$$E = \frac{1}{2}mv^2 = \frac{\hbar^2 k^2}{2m}$$

(Photon: E = $\hbar c k$

λ = 1.8 Å $\Rightarrow$ E $\approx$ 7 keV)

Thermal neutron: T = 293 K

$\Rightarrow$ v = 2200 m/s

$\Rightarrow$ λ = 2π/k = 1.8 Å (atomic distances)

$\Rightarrow$ E = 25 meV (molecular/lattice excitations)

Energy regimes

Energy – momentum relation :

$$\boldsymbol{E = 81.81 \cdot \lambda^{-2} = 2.07 \cdot k^2 = 5.23 \cdot v^2}$$

λ [Å]	1	2	4	6	10
E [meV]	80	20	5	2	1
T [K]	960	240	60	24	12
v [m/s]	3911	1956	978	620	437

Cold neutrons :	0.1-10 meV	D_2 at 25 K
Thermal neutrons :	10-100 meV	D_2O at room T
Hot neutrons :	100-500 meV	graphite at 2400 K

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Comparison

Energy – momentum relation :

$$\boldsymbol{E = 81.81 \cdot \lambda^{-2} = 2.07 \cdot k^2 = 5.23 \cdot v^2}$$

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E [meV]	80	20	5	2	1
T [K]	960	240	60	24	12
v [m/s]	3911	1956	978	620	437

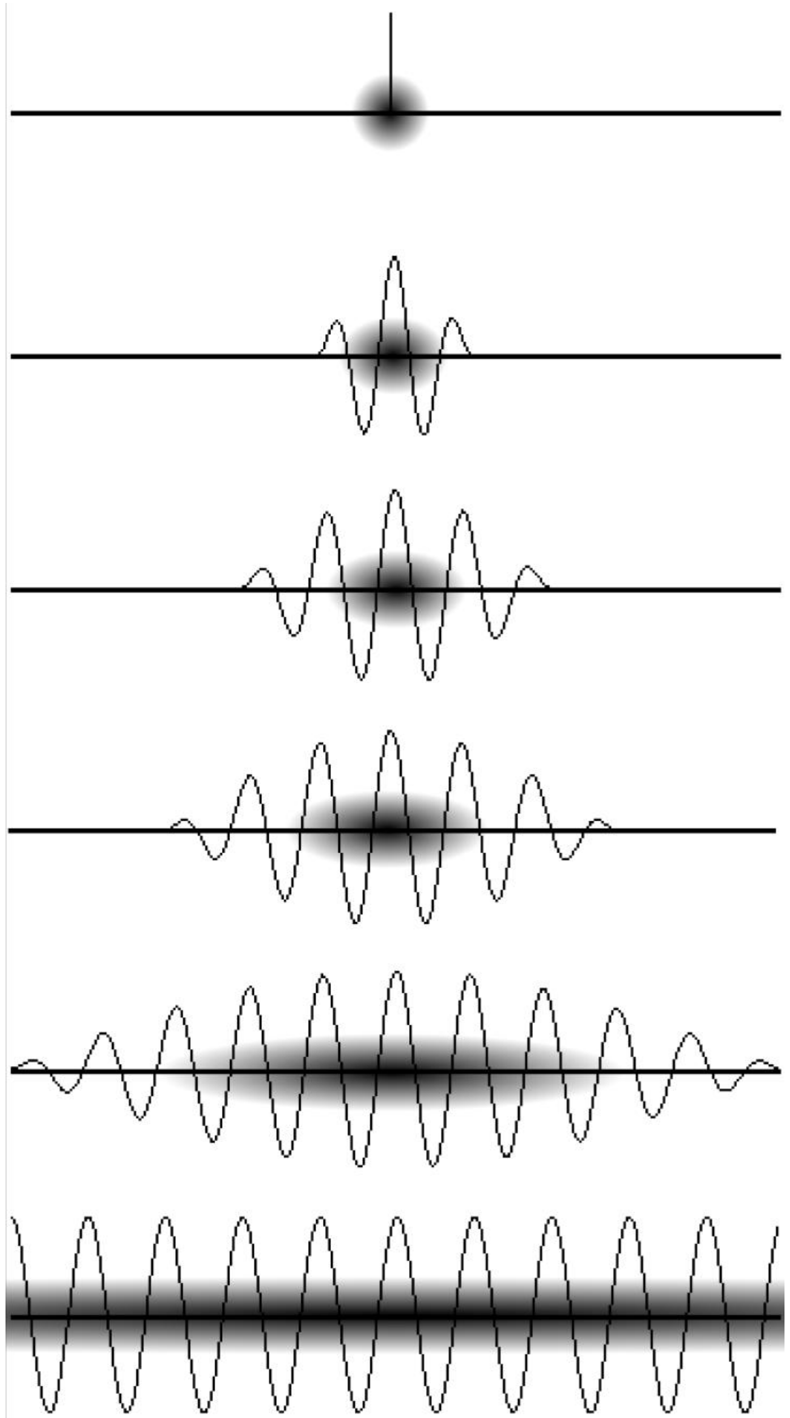
X – ray properties : $E = \hbar c k$, c = $3 \cdot 10^9$ m/s

Inelastic x-ray scattering : $\lambda = 1$ Å → $E = 12.4$ keV

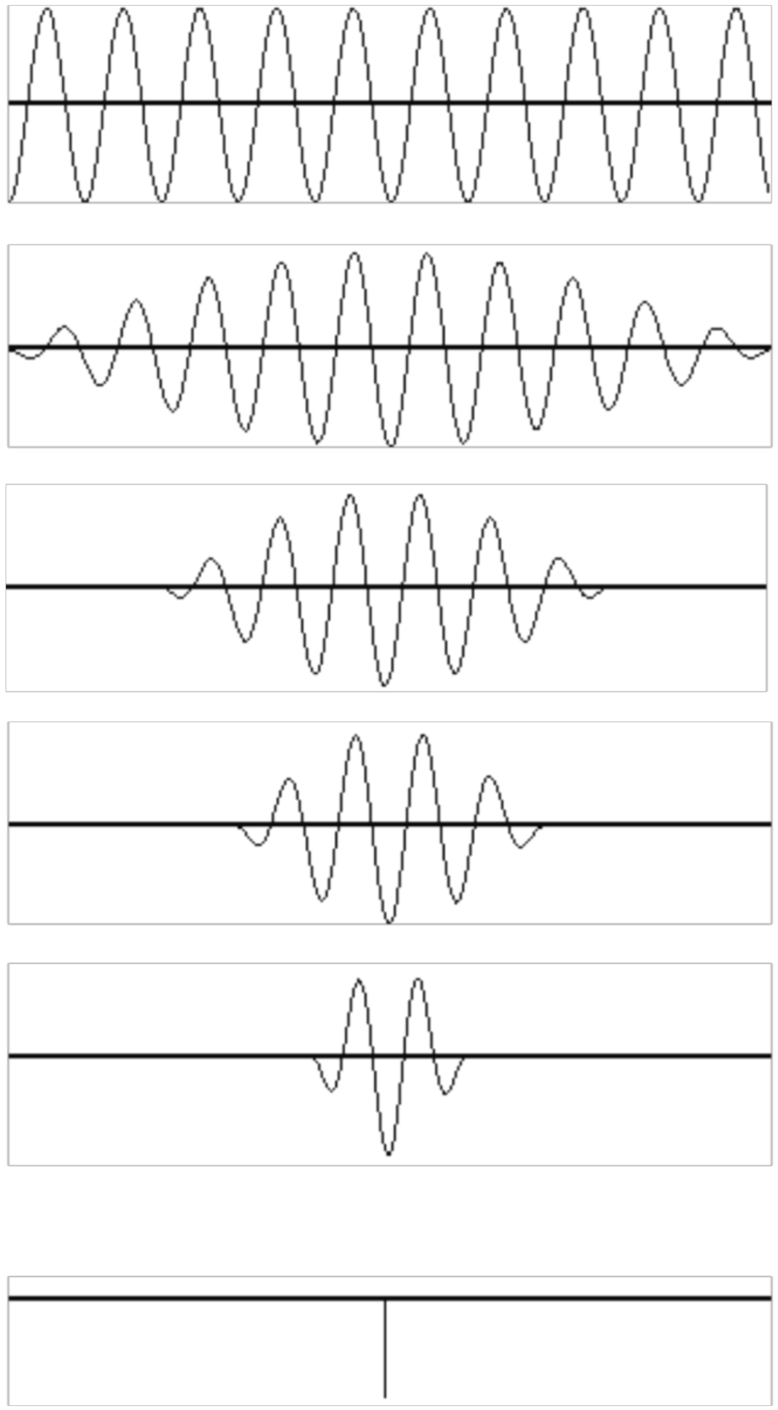
Raman, infra-red, ... : 10^3-10^5 Å → $E = 12.4 – 0.12$ eV

Heisenberg principle

Position



Momentum



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Heisenberg principle

Heisenberg's heritage : nothing is certain !

$$\Delta E \cdot \Delta t \geq \hbar\pi \qquad \Delta \nu \cdot \Delta t \geq ½ \qquad \Delta\omega \cdot \Delta t \geq \pi$$

$$\Delta p \cdot \Delta x \geq \hbar\pi \qquad \Delta\lambda^{-1} \cdot \Delta x \geq ½ \qquad \Delta k \cdot \Delta x \geq \pi$$

Let's get real : Spectrometer with $\Delta E/E$ of 5% (0.05%) utilized with neutrons of E = 2 meV - what can we measure ?

$$\Delta E = 0.1 \text{ meV} \rightarrow \Delta t \geq 20 \cdot 10^{-12} \text{ s} = 20 \text{ ps}$$

$$\Delta E = 1\ \mu\text{eV} \rightarrow \Delta t \geq 20 \cdot 10^{-14} \text{ s} = 2 \text{ ns}$$

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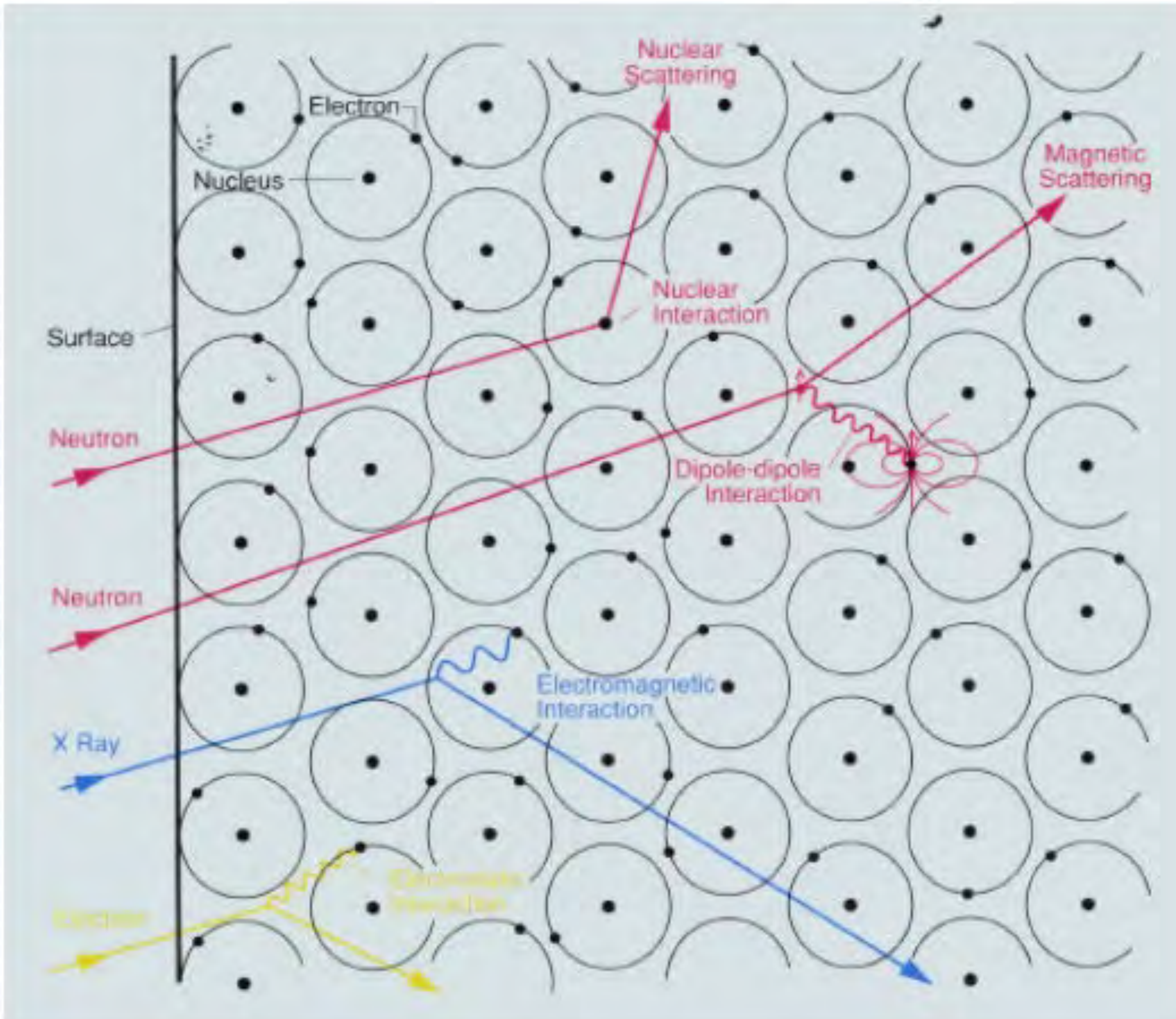
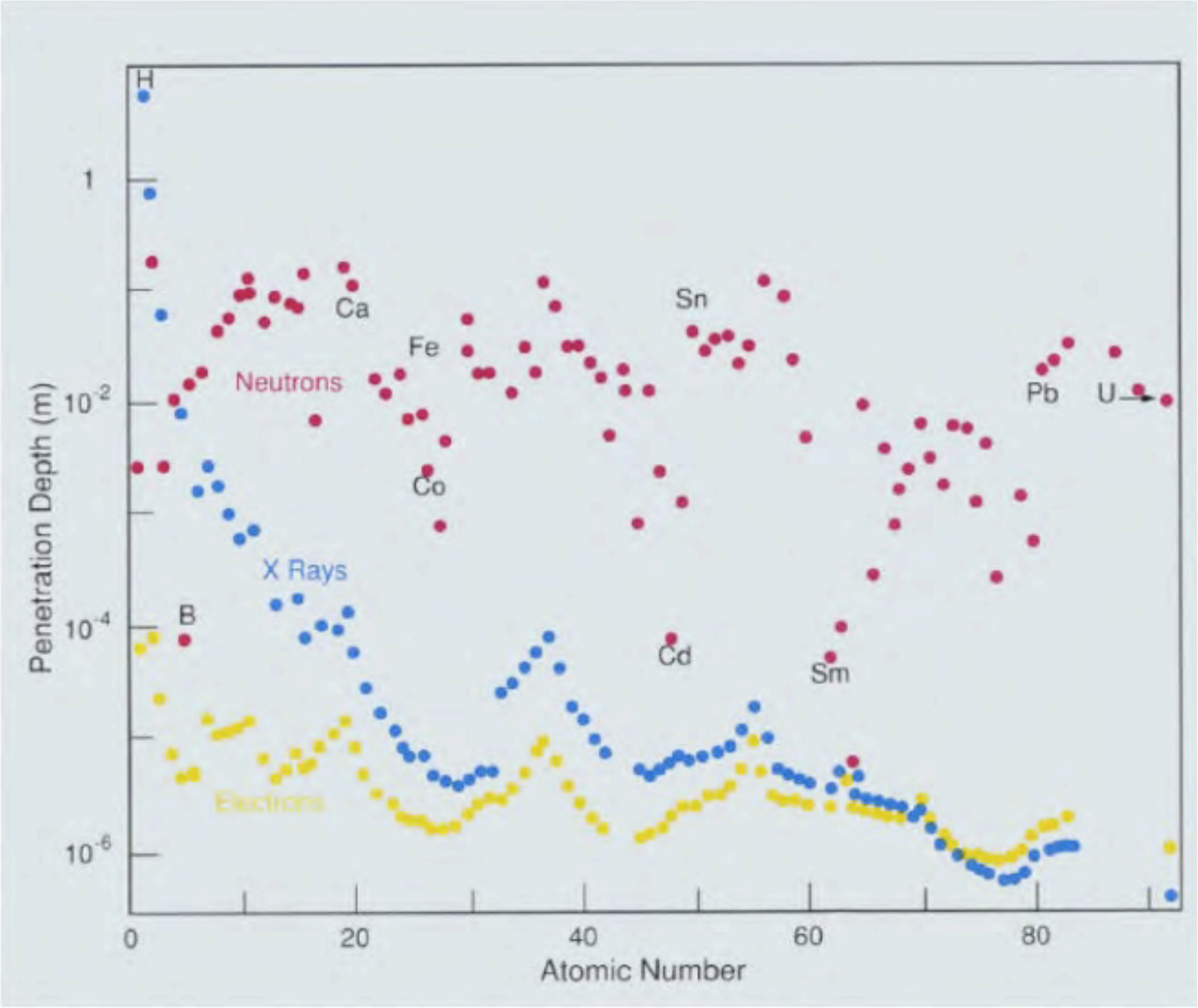
Interaction



Absorption



H
1
10^{-2}
10^{-4}
10^{-6}
Penetration Depth (m)
Ca
Fe
Neutrons
Co
X Rays
B
Sn
Cd
Sm
Pb
U
Electrons
0
20
40
60
80
Atomic Number

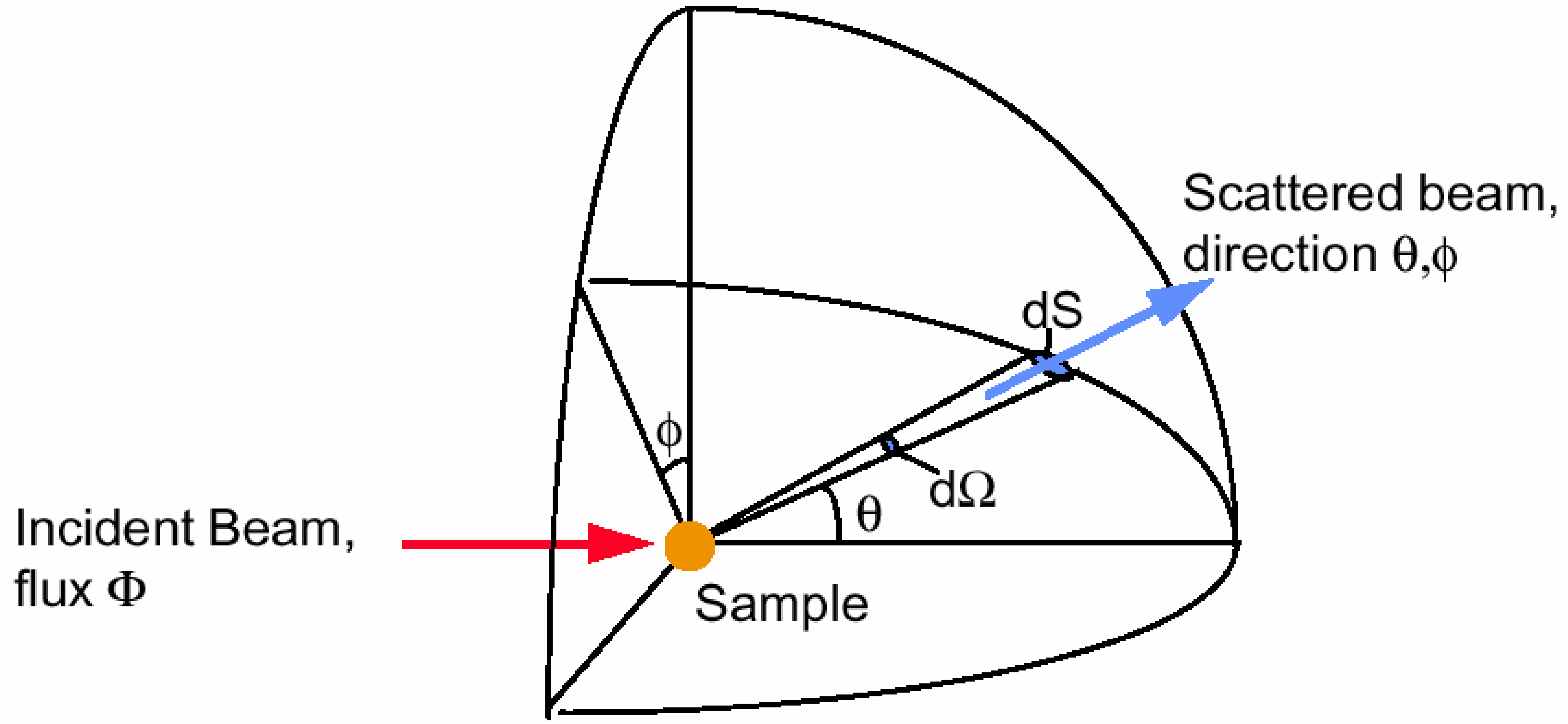
What can we measure?

Φ = number of incident particles per area and time

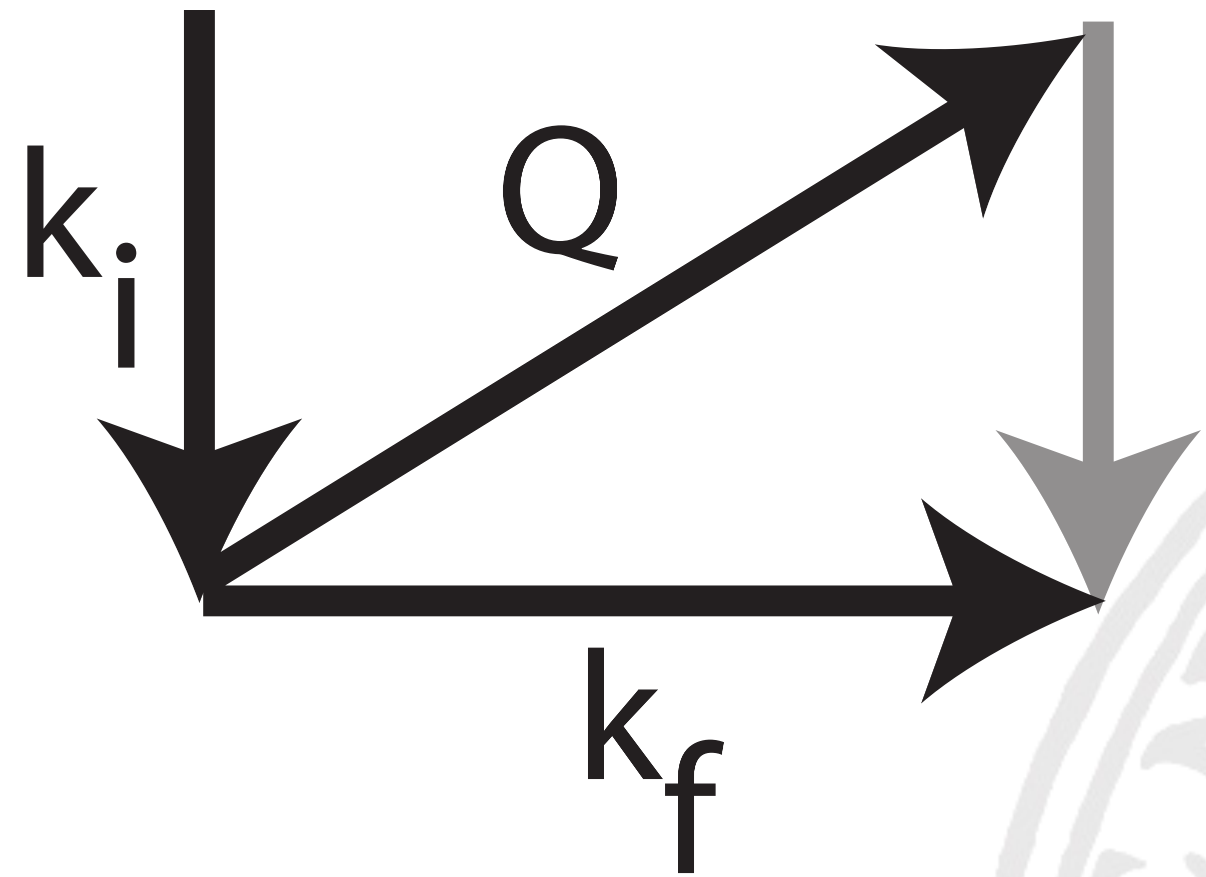
σ = total number of particles scattered per time and Φ

$d\sigma/d\Omega$ = number of particles scattered per time into a certain direction per area and Φ

$d^2\sigma/d\Omega dE$ = number of particles scattered per time into a certain direction per area with a certain energy per energy interval and Φ



Definitions



$$\vec{Q} = \vec{k}_f - \vec{k}_i$$

$$\omega = \frac{\hbar k_f^2}{2m} - \frac{\hbar k_i^2}{2m}$$

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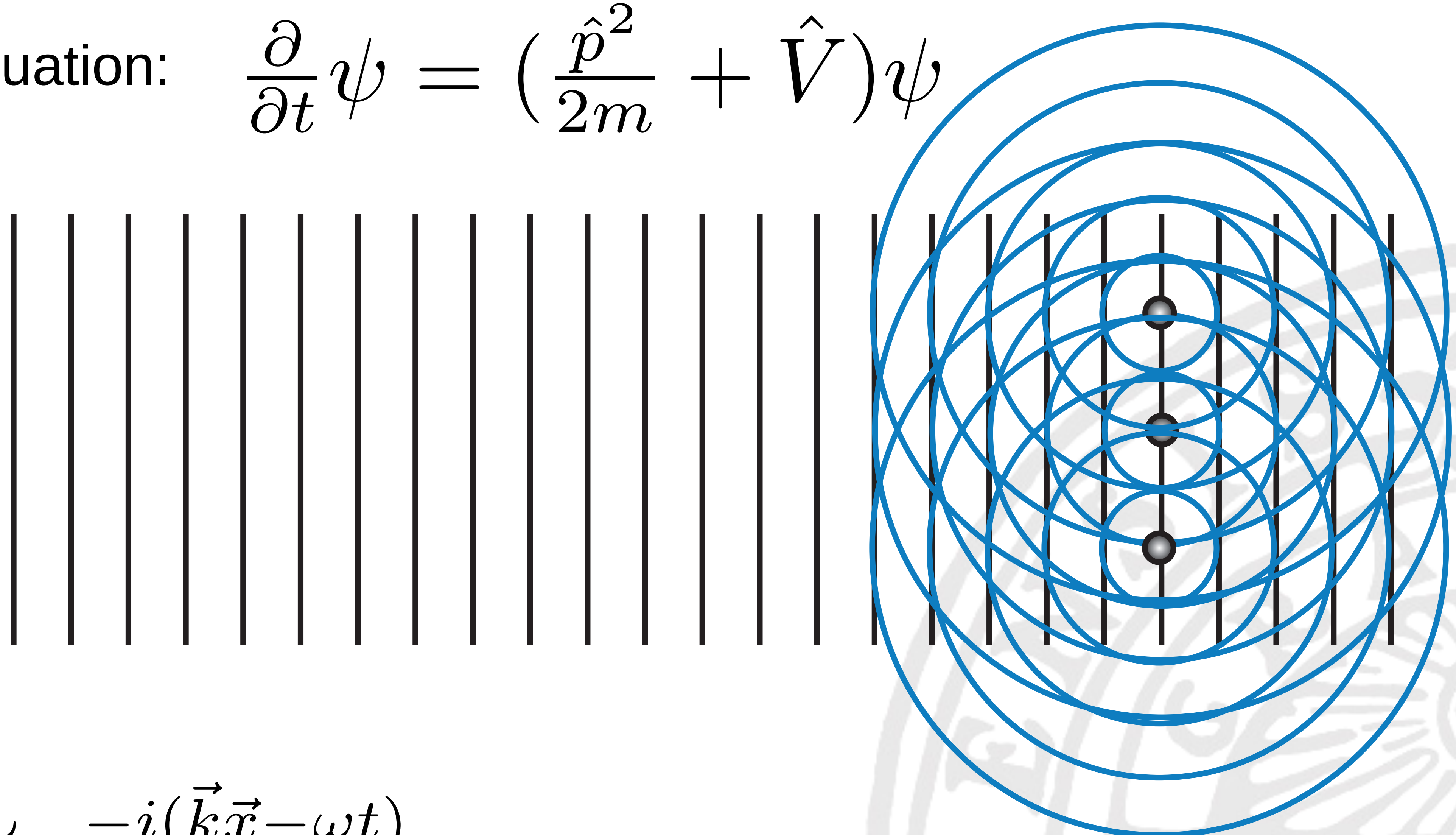
Mathematical description

Schrödinger equation: $$\frac{\partial}{\partial t}\psi = (\frac{\hat{p}^2}{2m} + \hat{V})\psi$$



$$\psi_i(\vec{r},\omega) \cong e^{-i(\vec{k}\vec{x}-\omega t)}$$

$$\psi_f(\vec{r},\omega) \cong e^{-i(\vec{k}\vec{x}-\omega t)} + f(\theta)\frac{e^{-i(\vec{k}\vec{r}-\omega t)}}{\vec{r}}$$

Kinematic approximation

Kinematic approximation means:

Combination of s-wave scattering (for neutrons)
with the
Born approximation (single event scattering)

The following effects are neglected (dynamic theory):

- Attenuation of the beam.
- Absorption.
- Primary extinction (attenuation due to scattering).
- Secondary extinction (attenuation due to multiple scattering).

We will also not consider the finite size of the sample (lattice sum).

All these effects are important for perfect crystals that hardly exist is disordered materials.

Scattering function

Scattering potential for neutrons: $V(\vec{r},t) \propto (b_n\delta(\vec{r}-\vec{R}) + b_m(\vec{r}-\vec{R}))$

The scattering function: $$S(\vec{Q},\omega) = \frac{4\pi}{\sigma_s}\frac{k_i}{k_f}\frac{\partial^2\sigma}{\partial\Omega\partial\omega}$$

$$S(\vec{Q},\omega) \cong \sum_{\alpha_f,\alpha_i} \rho(E_f)|\langle k_f,\omega_f,\hat{\sigma}_f|\hat{V}|\hat{\sigma}_i,\omega_i,k_i\rangle|^2\delta(\omega-\omega_{\alpha_f,\alpha_i})$$

This equation is also known as scattering law or dynamical structure factor. It **completely describes the structural and dynamic properties** of the sample.

$$\sigma_s = \int\int \frac{\partial^2\sigma}{\partial\Omega\partial\omega}d\Omega d\omega = 4\pi b^2$$

Potential

For neutrons (scattering at the core) wave length (10^{-10} m) is much larger than the size of the scattering particle (10^{-15} m). This implies that no details of the core can be seen and the scattering potential can be described by a single constant b, the scattering length:

$$V(\vec{r}) = \frac{2\pi h}{m} b\delta(\vec{r} - \vec{R})$$

This is called the Fermi pseudo potential. B is a phenomenological constant and has to be determined experimentally.

Cross section

The Fermi pseudo potential is true neutron-nucleus potential, but it is constructed to provide isotropic s-wave scattering in the Born approximation.

Inserting this potential into the scattering cross section results in:

$$\frac{\partial\sigma}{\partial\Omega} = b^2$$

The total scattering cross section is then:

$$\sigma_{tot} = \int \frac{\partial\sigma}{\partial\Omega} d\Omega = 4\pi b^2$$

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Ensemble

The potential for an ensemble of scatterers can be described by the sum of the scattering length:

$$V(\vec{r}) = \frac{2\pi h}{m} \sum_i b_i \delta(\vec{r} - \vec{R}_i)$$

The sum is taken over all nuclei at positions R_i.

For the cross section this results in:

$$\frac{\partial \sigma}{\partial \Omega} = \left| \sum_i b_i e^{i\vec{Q}\vec{r}} \right|^2 = \sum_{i,j} b_i b_j e^{i\vec{Q}\left(\vec{R}_i - \vec{R}_j\right)} = \sum_{ij} b_i b_j e^{i\vec{Q}\vec{R_{ij}}}$$

R_{ij} is the distance between the scatterers or the lattice parameter for a crystal.

Average

In a scattering experiment the ensemble average or time average is measured:

$$\frac{\partial\sigma}{\partial\Omega} = \left\langle \sum_{ij} b_i b_j e^{i\vec{Q}\vec{r}} \right\rangle$$

Assuming that the scatterers are fixed at their positions the average has to be taken only over the scattering length:

$$\frac{\partial\sigma}{\partial\Omega} = \sum_{ij} \langle b_i b_j \rangle \, e^{i\vec{Q}\vec{R}_{ij}}$$

Formally we can divide up the double sum into to sums:

$$\frac{\partial\sigma}{\partial\Omega} = \sum_{i=j} \langle b_i b_j \rangle \, e^{i\vec{Q}\vec{R}_{ij}} + \sum_{i\neq j} \langle b_i b_j \rangle \, e^{i\vec{Q}\vec{R}_{ij}}$$

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Averaging

$$b_i = \langle b_i \rangle + \delta b_i = b_i$$

As δb_i averages to 0.

$$b_i b_j = \langle b_i \rangle \langle b_j \rangle + \underbrace{\langle b_i \rangle \delta b_j}_{=0} + \underbrace{\langle b_j \rangle \delta b_i}_{=0} + \delta b_j \delta b_i = \langle b^2 \rangle - \langle b \rangle^2$$

For no correlations i=j.

Inserting into the cross section gives:

$$\frac{d\sigma}{d\Omega} = \underbrace{\langle b \rangle^2 \sum_{ij} \exp\left(i\vec{K}\cdot\vec{R}_{ij}\right)}_{\left(\frac{d\sigma}{d\Omega}\right)_{coh}} + \underbrace{N\left(\langle b^2 \rangle - \langle b \rangle^2\right)}_{\left(\frac{d\sigma}{d\Omega}\right)_{incoh}}$$

$b_{coh}=\langle b\rangle$ and $b_{inc}=(\langle b^2\rangle-\langle b\rangle^2)^{1/2}$ is then called coherent and incoherent scattering length, respectively.

Spin incoherent scattering

For a one atom with nuclear angular moment J the total moment of the neutron nucleus pair has to be considered.

The neutron is a Spin ½ particle. In an unpolarised neutron beam S= ½ and S=- ½ neutrons are present.

For the total moment of the system we get: M=J±S.

Accordingly two scattering length have to be considered:

b_+ for M=J+ ½

and

b_- for M=J- ½

Generally, 2J+1 different values are possible for the angular moment J. This results in a total number of 2(2J+1) eigenstates (2(J+1) for b_+ and 2J for b_-).

Spin incoherent scattering

The probability for measuring b_+ is then:

$$p_+ = \frac{2(J+1)}{2(2J+1)} = \frac{J+1}{2J+1}$$

And for b_-:

$$p_- = \frac{2(J+1)}{2(2J+1)} = \frac{J}{2J+1}$$

For J=0 we get 1 for b_+ and 0 for b_-.

The coherent and incoherent scattering length can then be calculated:

$$b_{coh}^2 = \langle b \rangle^2 = \left(p_+ b_+ + p_- b_- \right)^2$$

$$b_{inc}^2 = \left(\langle b^2 \rangle - \langle b \rangle^2 \right) = p_+ p_- (b_+ - b_-)^2$$

Example ^1H

	I	Scattering lengths
^1H	1/2	$b_+ = 10.8$ fm $b_- = 47.4$ fm

This results in: $p_+=3/4$ and $p_-=1/4$.

$$b^2_{coh} = 14.06\ \text{fm}^2, \quad \sigma_{coh} = 4\pi b^2{}_{coh} = 1.76 barn$$

$$b^2_{inc} = 635.1\ \text{fm}^2, \quad \sigma_{inc} = 4\pi b^2{}_{inc} = 79.8 barn$$

Hydrogen scatters mainly incoherent!

Example ^2H

	I	Scattering lengths
^2H	1	$b_+ = 9.52$ fm $b_- = 0.96$ fm

This results in: $p_+=2/3$ and $p_-=1/3$.

$$b^2_{coh} = 44.5\ \mathrm{fm}^2, \quad \sigma_{coh} = 4\pi b^2{}_{coh} = 5.59 barn$$

$$b^2_{inc} = 16.32\ \ \mathrm{fm}^2, \ \sigma_{inc} = 4\pi b^2{}_{inc} = 2.05 barn$$

Deuterium scatters mainly coherent!

Correlation functions

Coherent scattering: $$S_{coh}(Q,\omega) = \int K(r,t)e^{i(Qr-\omega t)}drdt$$

$\Rightarrow$ Bragg reflections, liquid structure factor, phonon scattering etc.

Example (Bragg law): $$n\lambda = 2d_{hkl}\sin(\phi/2) \qquad d_{hkl} = \frac{2\pi}{Q}$$

Incoherent scattering: $$S_{inc}(Q,\omega) = \int K_s(r,t)e^{i(Qr-\omega t)}drdt$$

$\Rightarrow$ Scatterers exist, self motion etc.

Example

(Random jump diffusion):

$$K(r,t) = e^{-Dtr^2}$$

$$S(Q,\omega) = \frac{1}{\pi}\frac{DQ^2}{\omega^2 + (DQ^2)^2}$$

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Real - reciprocal space

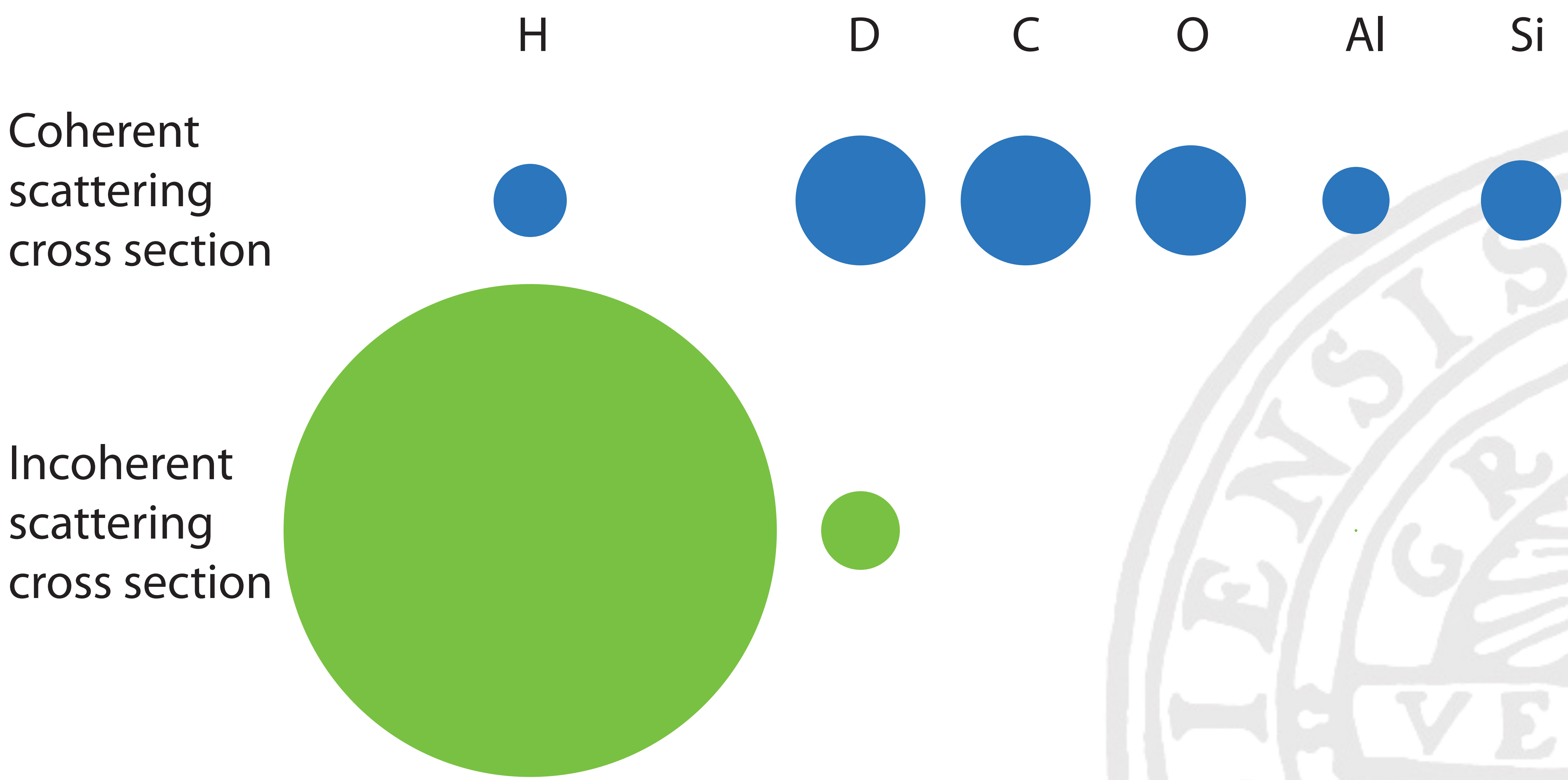
Observable in INS and Diff.

$$
\begin{tikzcd}
\boxed{S(\vec{Q},\omega)} \arrow[r, leftrightarrow] \arrow[d, leftrightarrow] & J(\vec{r},\omega) \arrow[d, leftrightarrow] \\
\boxed{I(\vec{Q},t)} \arrow[r, leftrightarrow] & \boxed{G(\vec{r},t)}
\end{tikzcd}
$$

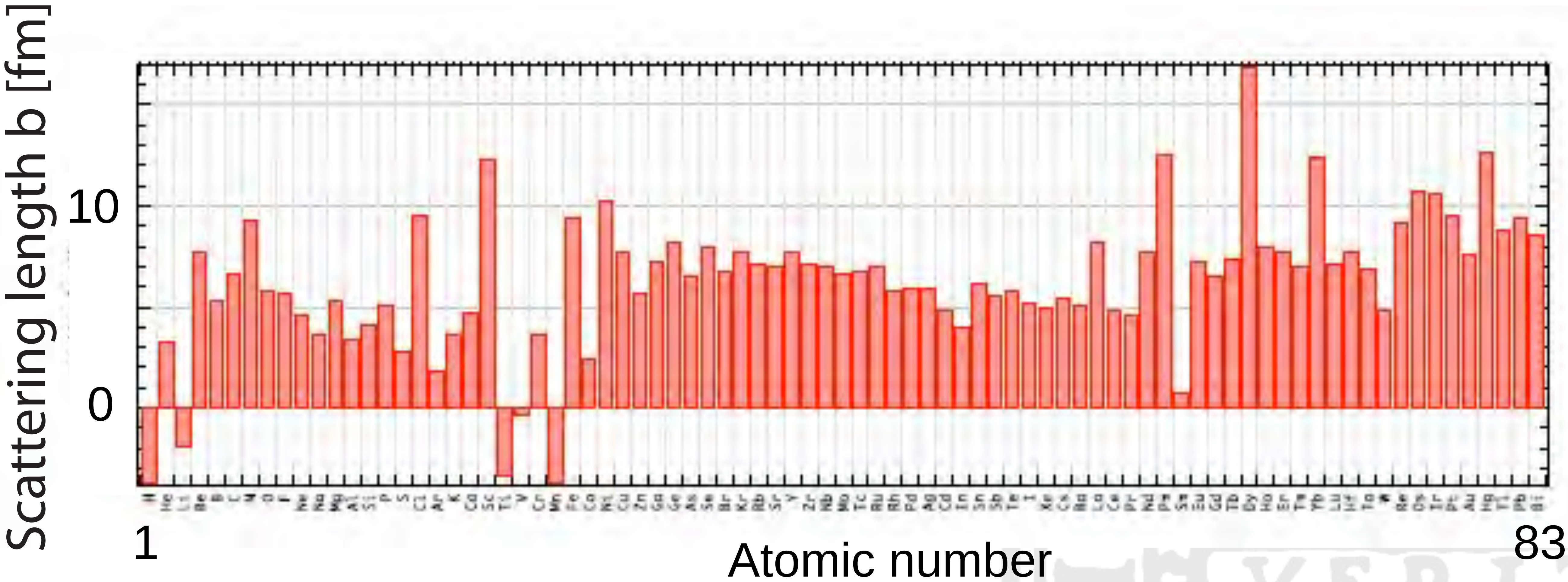
Observable in NSE,
simple models

Simple models,
computer simulations

Scattering cross section



Scattering length



Summary

Neutrons:

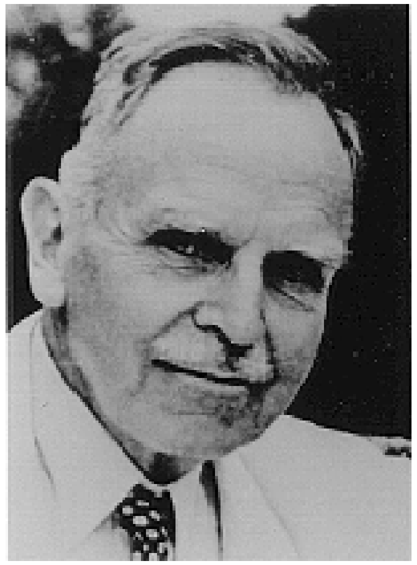
Weakly interacting → High penetration
Spin → Magnetic sensitivity
Core interaction → Isotope sensitivity
Coherent/Incoherent scattering
Low energy → Good energy resolution

Scattering function describes a sample completely

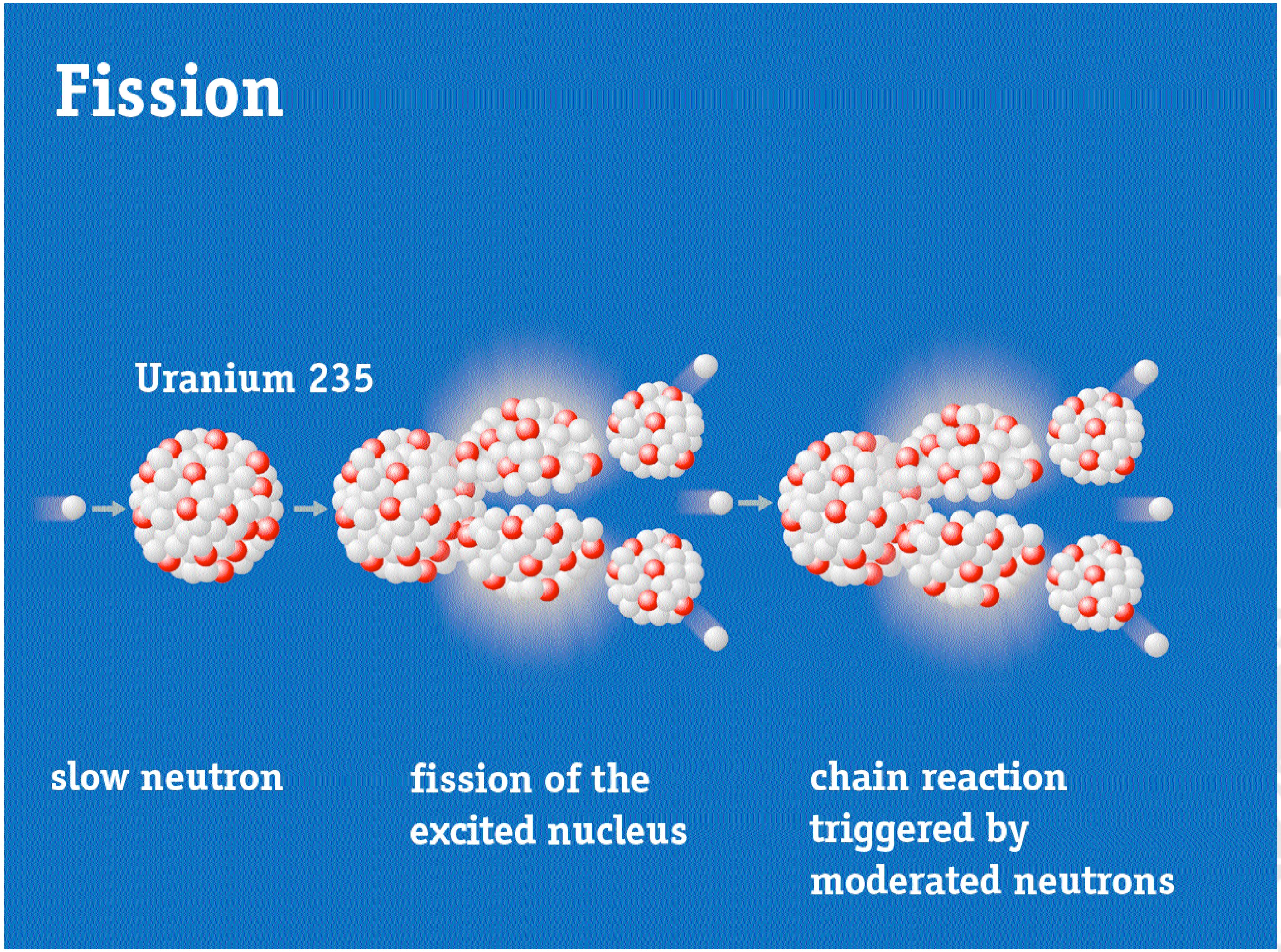
Instrumentation:

Time of flight
Angle dispersive

Nuclear reactor

Lise Meitner und Otto Hahn



... discovered 1938 together with Fritz Strassmann the nuclear fission reaction of Uranium after irradiation with neutrons

$$n + {}^{235}_{92}U \rightarrow {}^{236}_{92}U \rightarrow {}^{144}_{56}Ba + {}^{89}_{36}Kr + 3n + 177MeV$$

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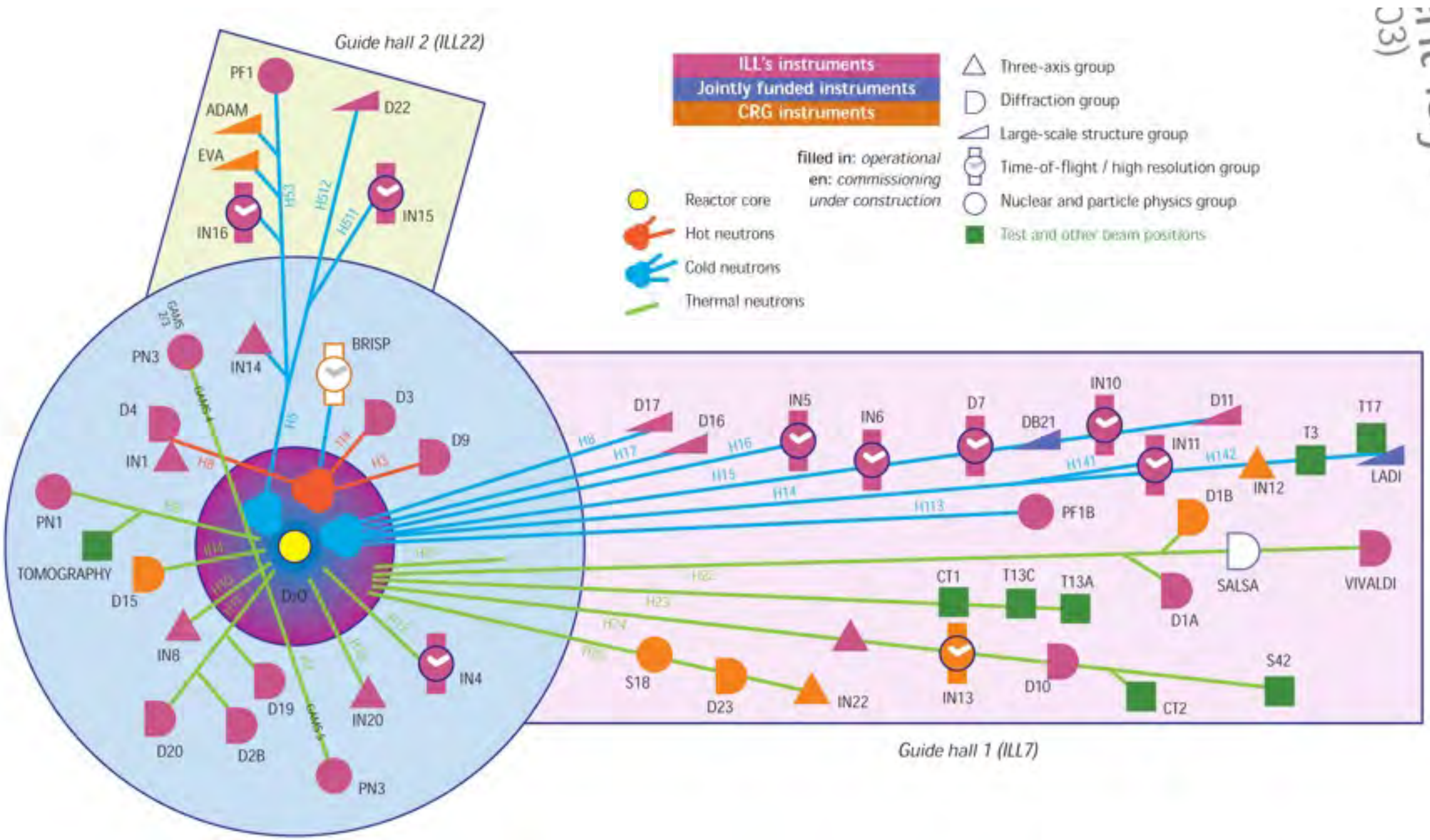
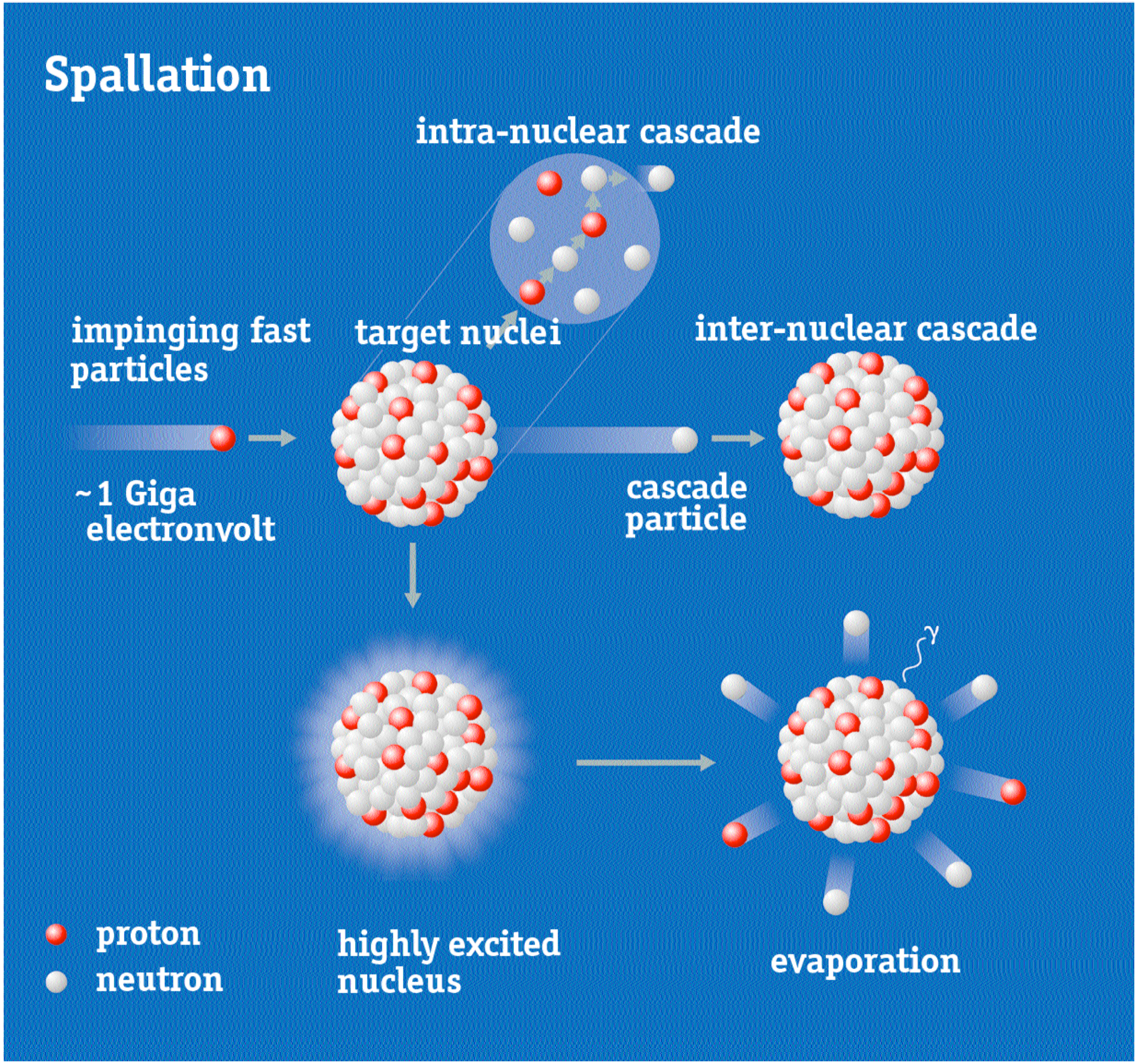
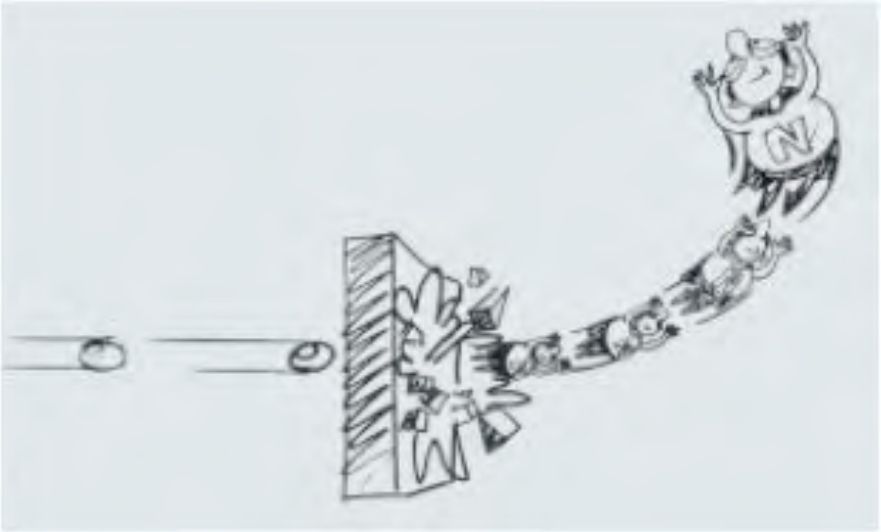
Floor plan



Spallation



Spallation
intra-nuclear cascade
impinging fast particles
target nuclei
inter-nuclear cascade
~1 Giga electronvolt
cascade particle
γ
proton
neutron
highly excited nucleus
evaporation



N

ESS - How it will look like

MAX IV

Target station:

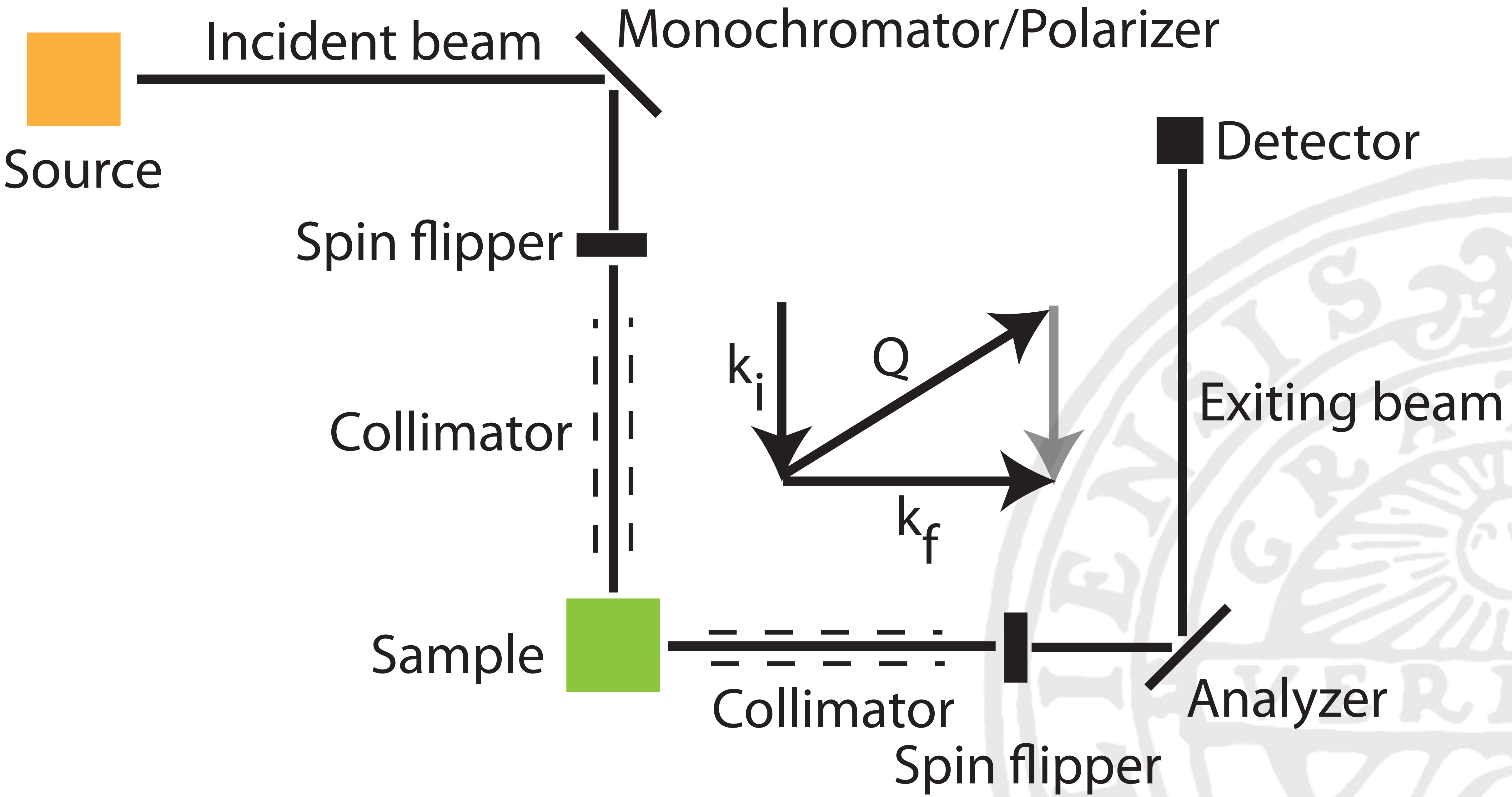
SP: 50 Hz
LP: 16.6 Hz
Pulse length 2 ms

Instrumentation

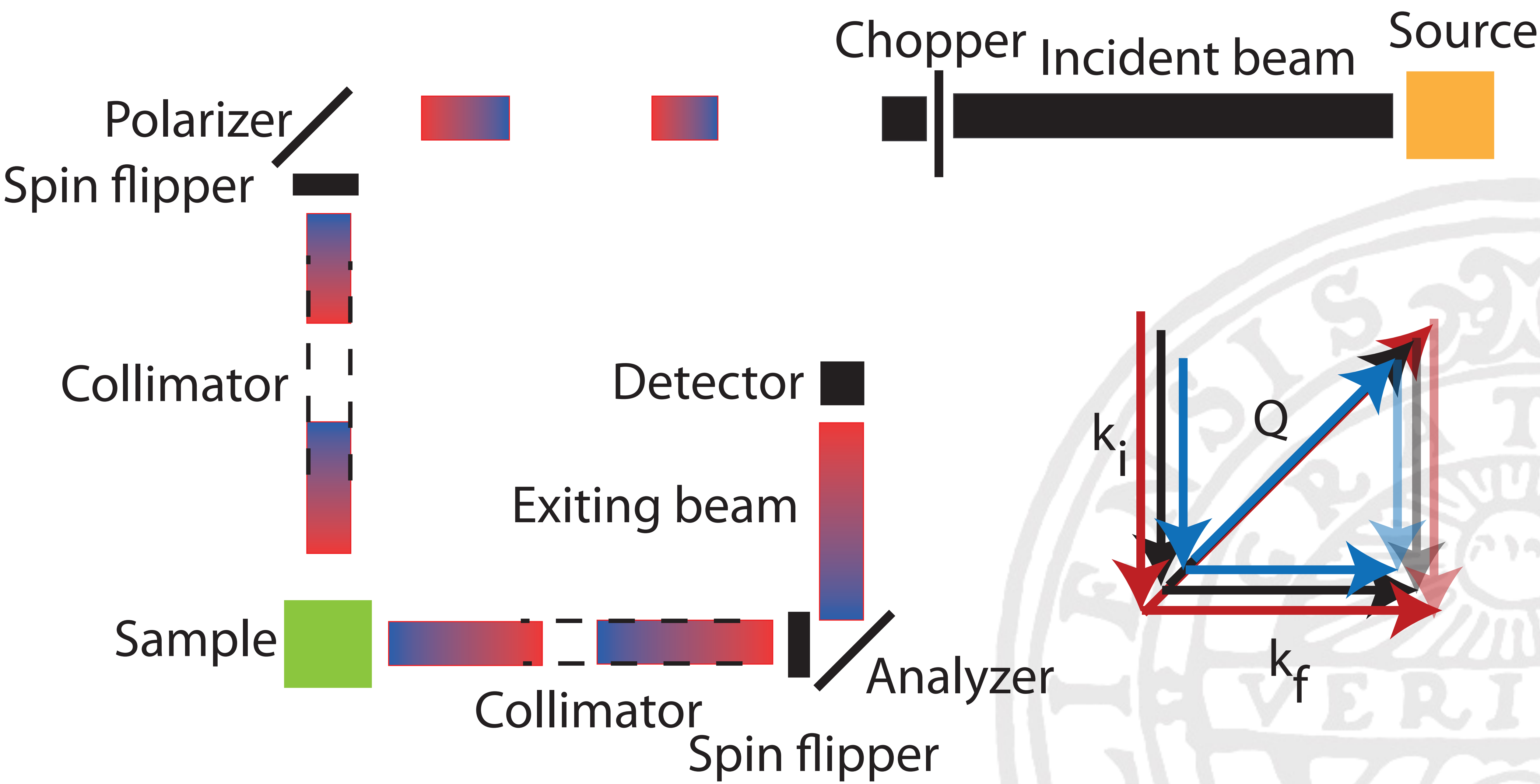
Linear accelerator:

NC: 769 m
SC: 432 m
Energy: 1.334 GeV
Current: 3.75 mA
Power: 5 MW

Angle dispersive neutron scattering

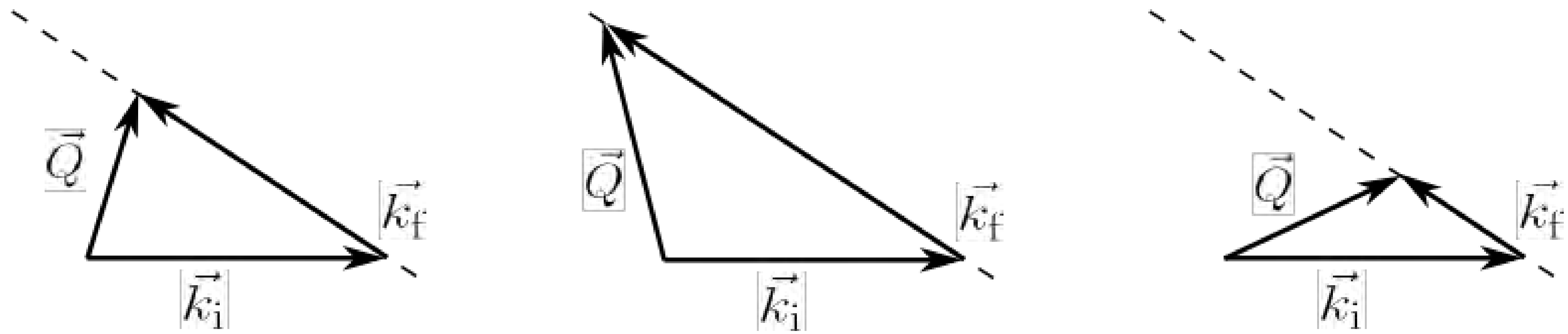


TOF neutron scattering

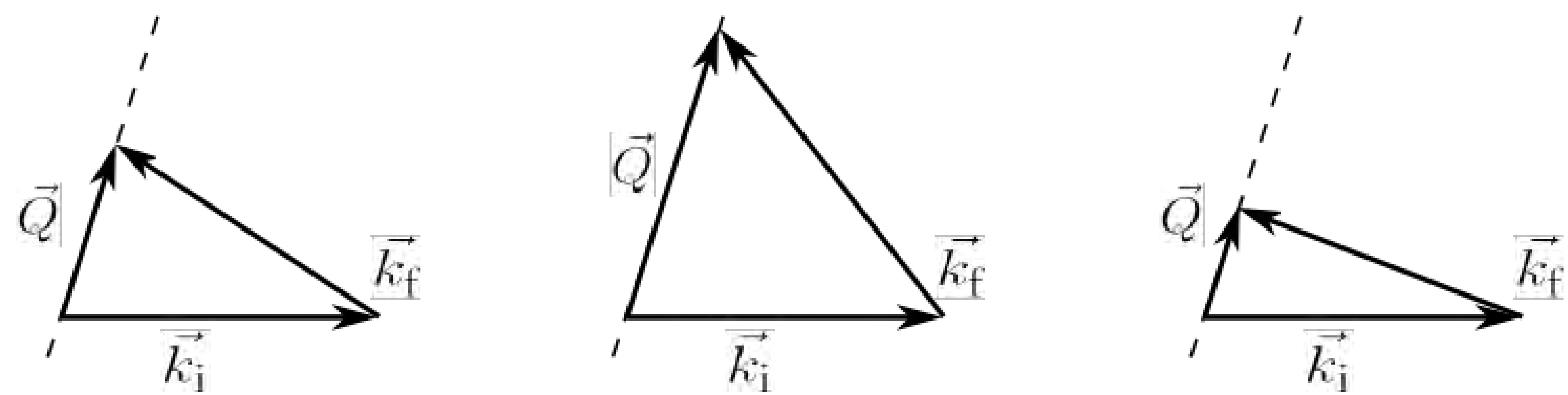


Scattering geometry

Elastic scattering	Neutron energy gain	Neutron energy loss
Rayleigh process	Anti-Stokes process	Stokes process



→ Constant directions of k_i and k_f but changing Q !



→ Constant directions of k_i and Q but changing k_f !

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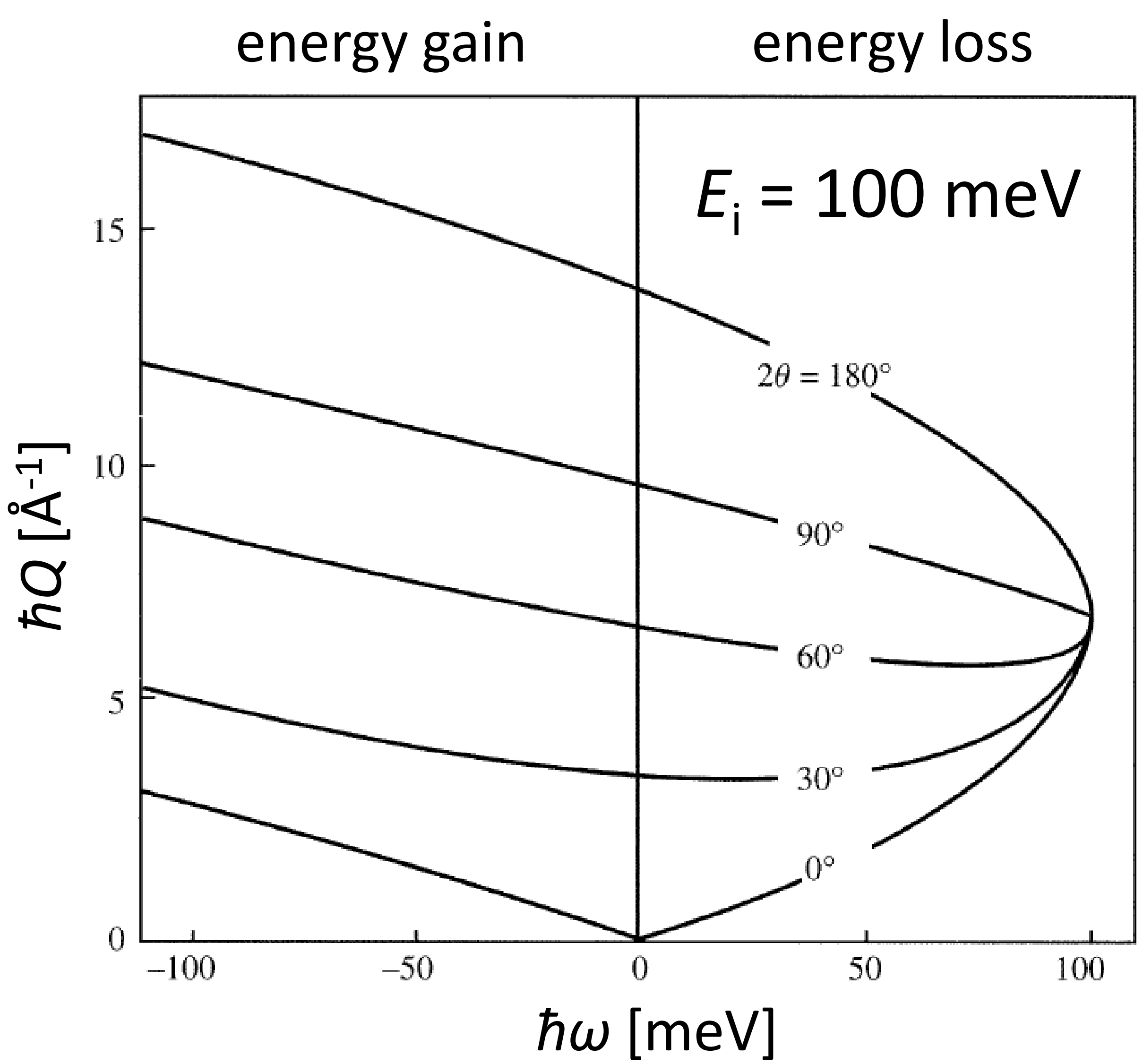
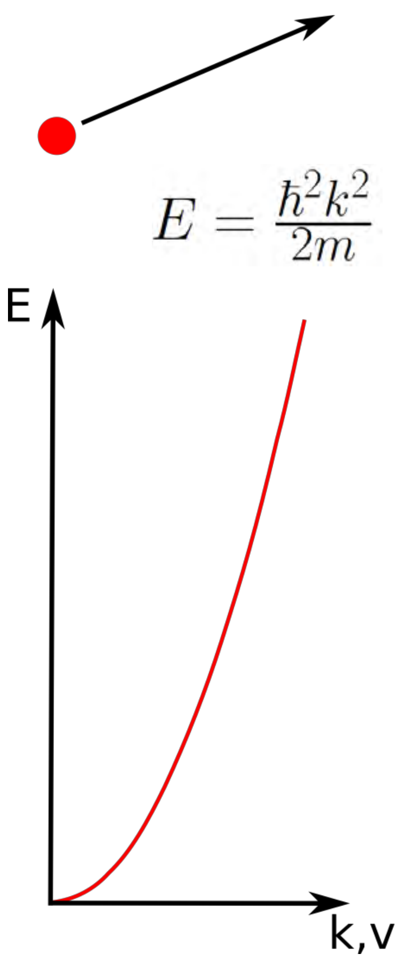
Kinematics

Kinematics of inelastically scattered neutrons :

$$\vec{Q}^2 = \vec{k}_\mathrm{i}^{\,2} + \vec{k}_\mathrm{f}^{\,2} - 2|\vec{k}_\mathrm{i}||\vec{k}_\mathrm{f}|\cos(2\Theta) \qquad \hbar\omega = E_\mathrm{i} - E_\mathrm{f} = \frac{\hbar^2}{2m_\mathrm{n}}(\vec{k}_\mathrm{i}^{\,2} - \vec{k}_\mathrm{f}^{\,2})$$



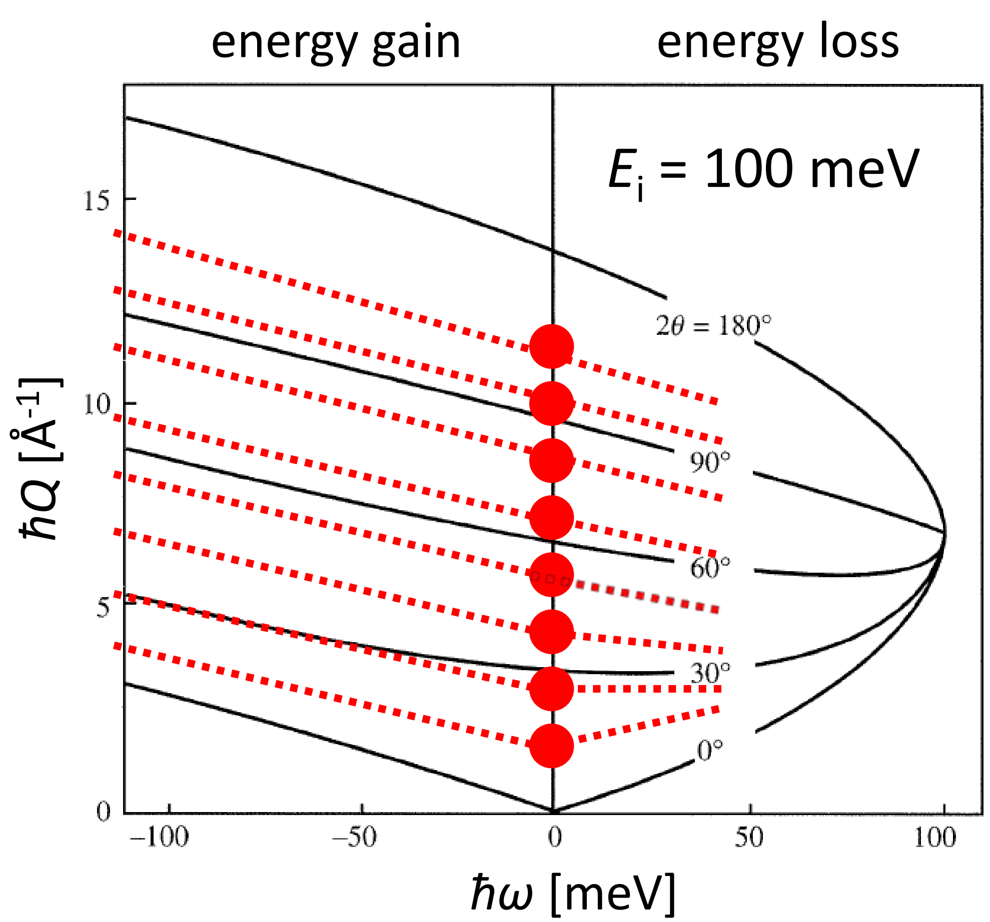


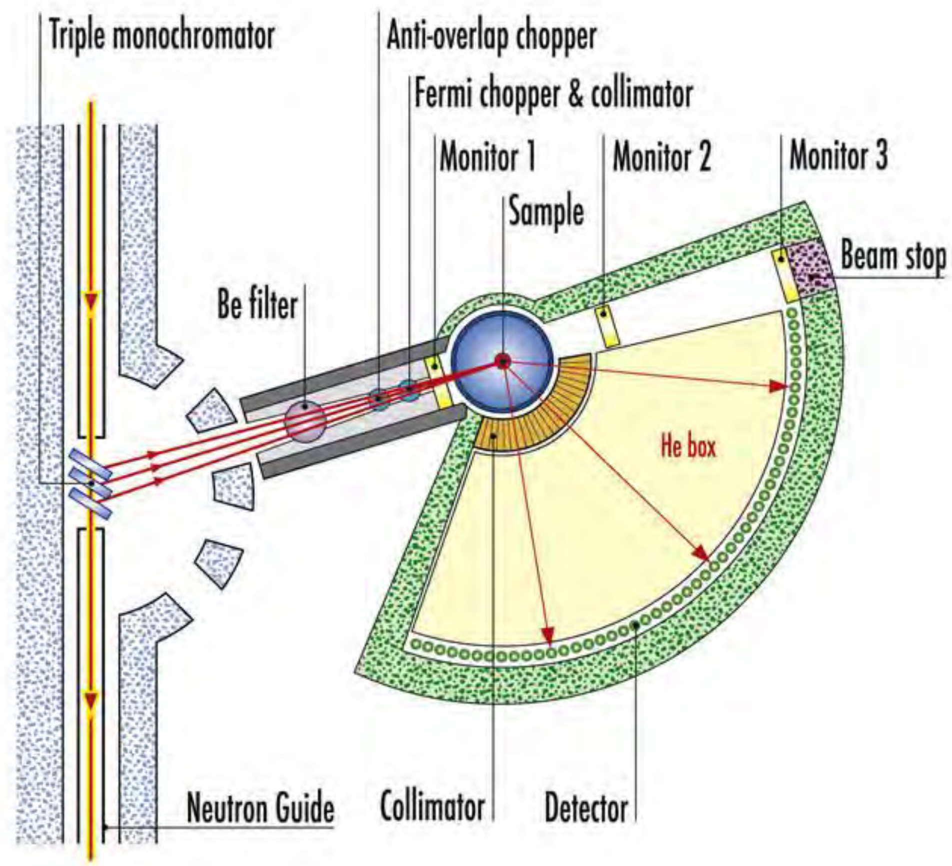
For 2θ = 0, 180°
$Q = \Delta k$, $E \sim \Delta k^2$

B.T.M. Willis & C.J. Carlile, ‘Experimental Neutron Scattering’, Oxford University Press:

Time of flight

Inelastic scattering instruments : Time-of-Flight Spectrometer

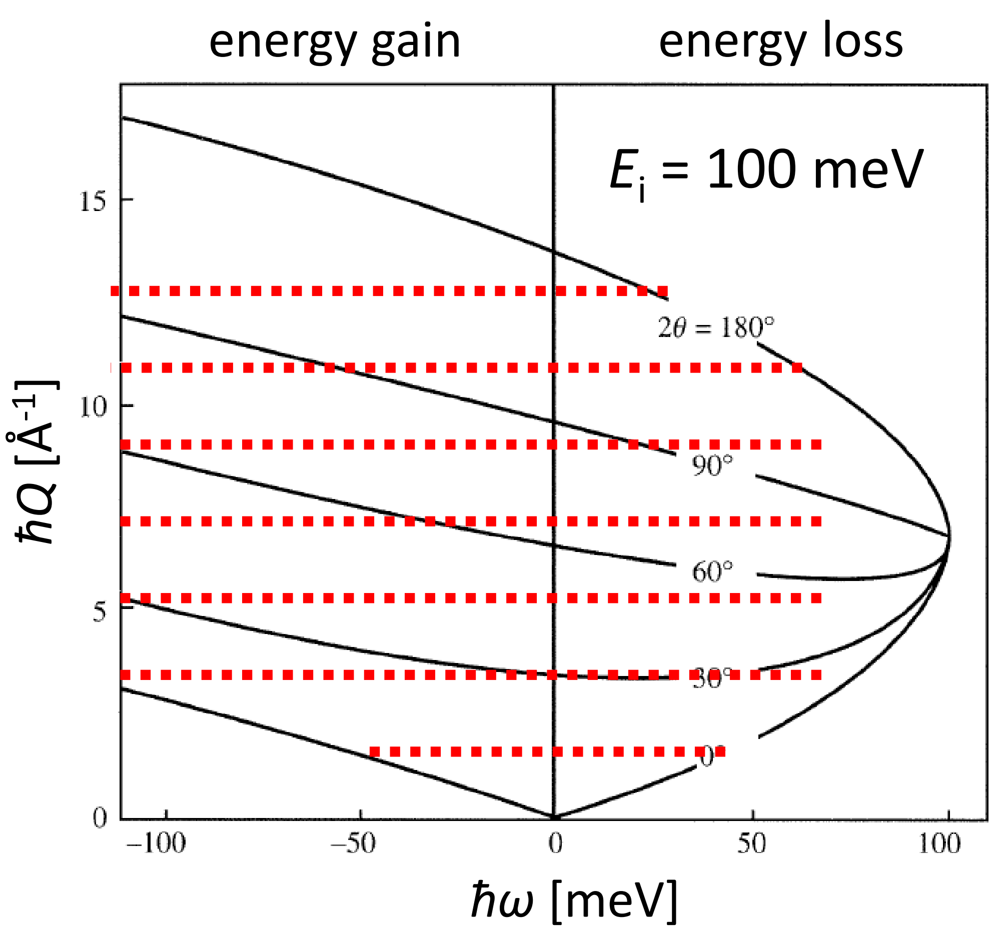


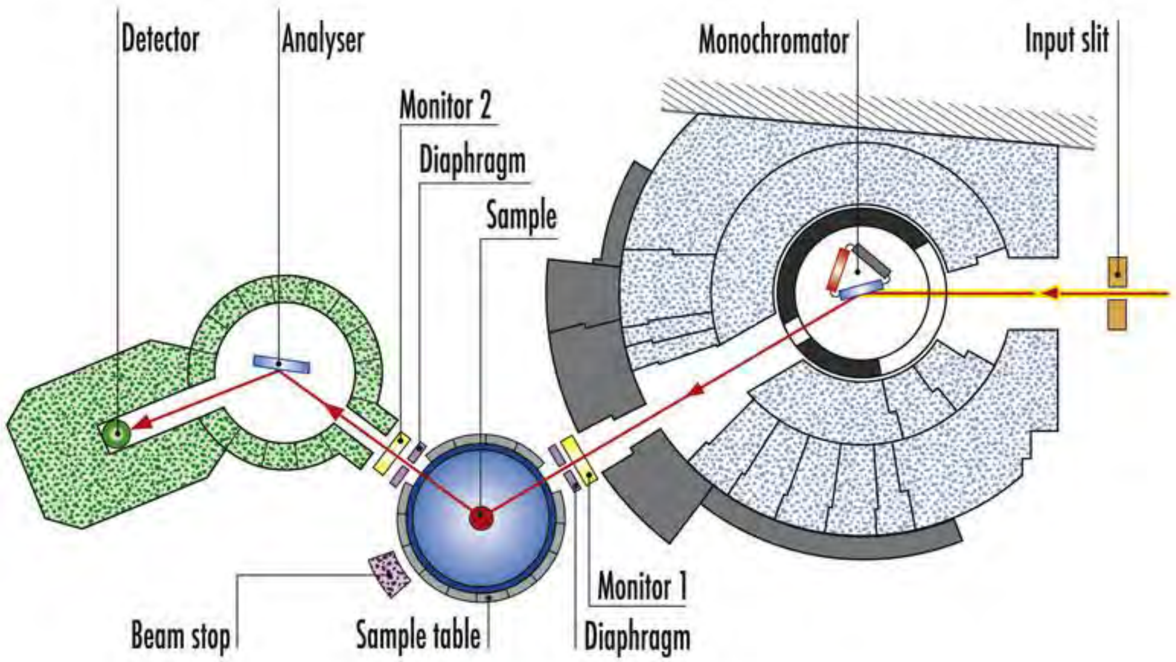


Equipped with large detector arrays to cover a wide 2θ range with typically 1 detector tube per degree of 2θ. Data are to be interpolated to constant *Q*.

Three axis

Inelastic scattering instruments : Three-Axis Spectrometer

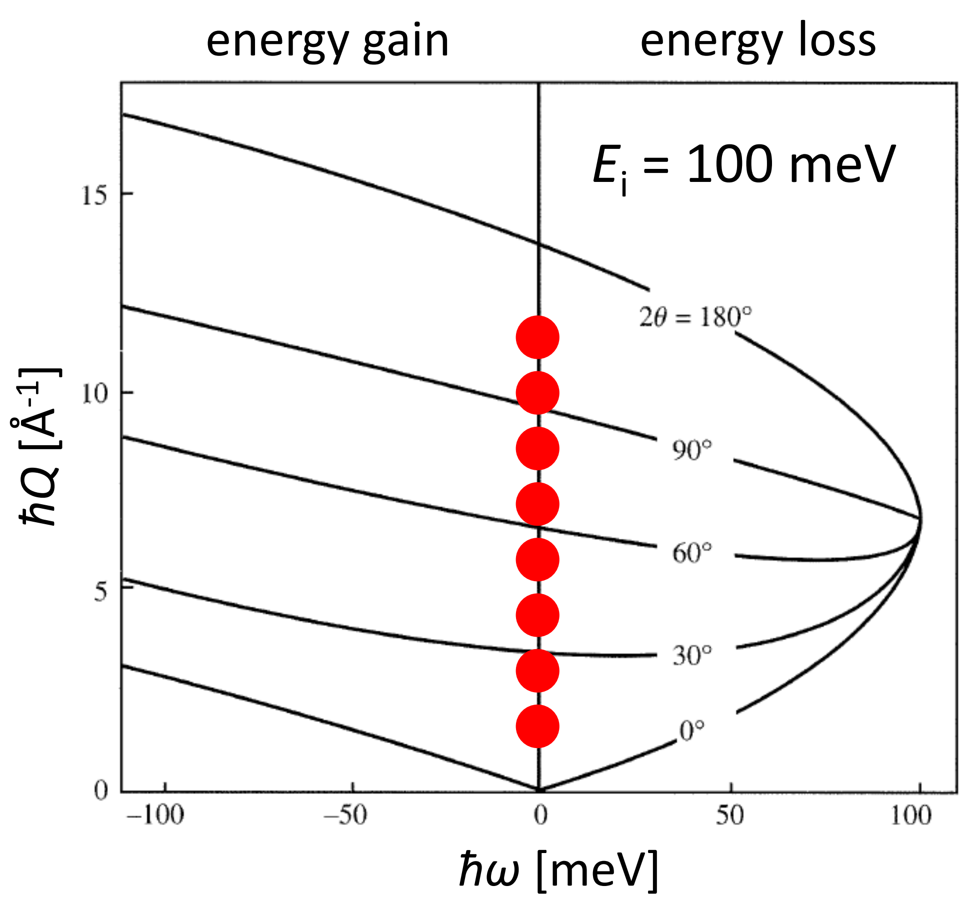


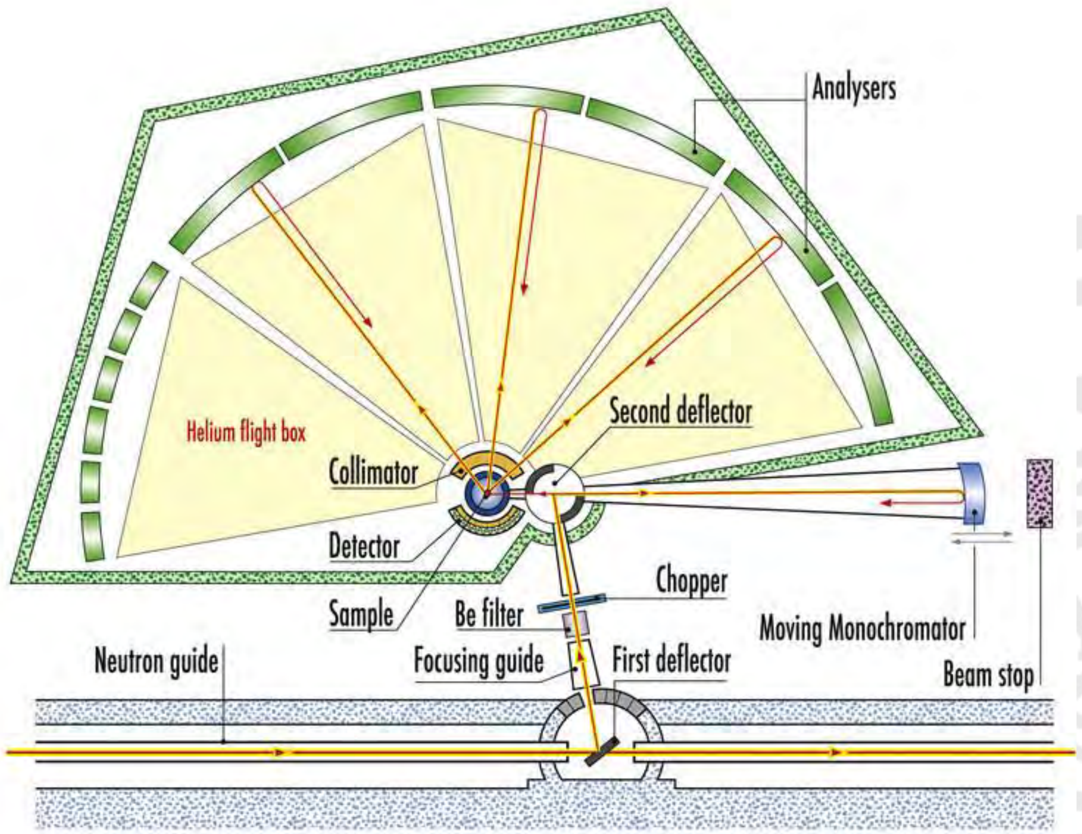


Typically equipped with a single detector but with highly flexible positioning capabilities to map out, e.g., constant *Q* slices.

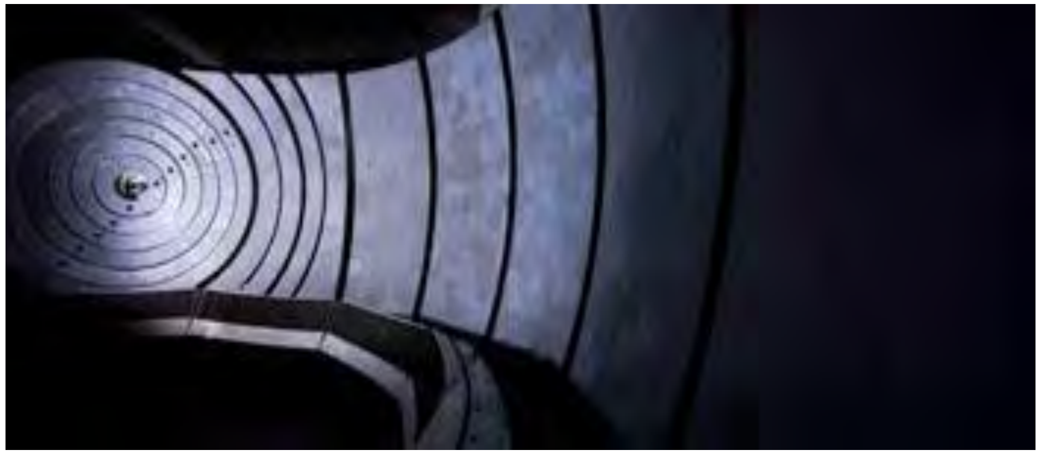
Backscattering

Inelastic scattering instruments : Back-Scattering Spectrometer





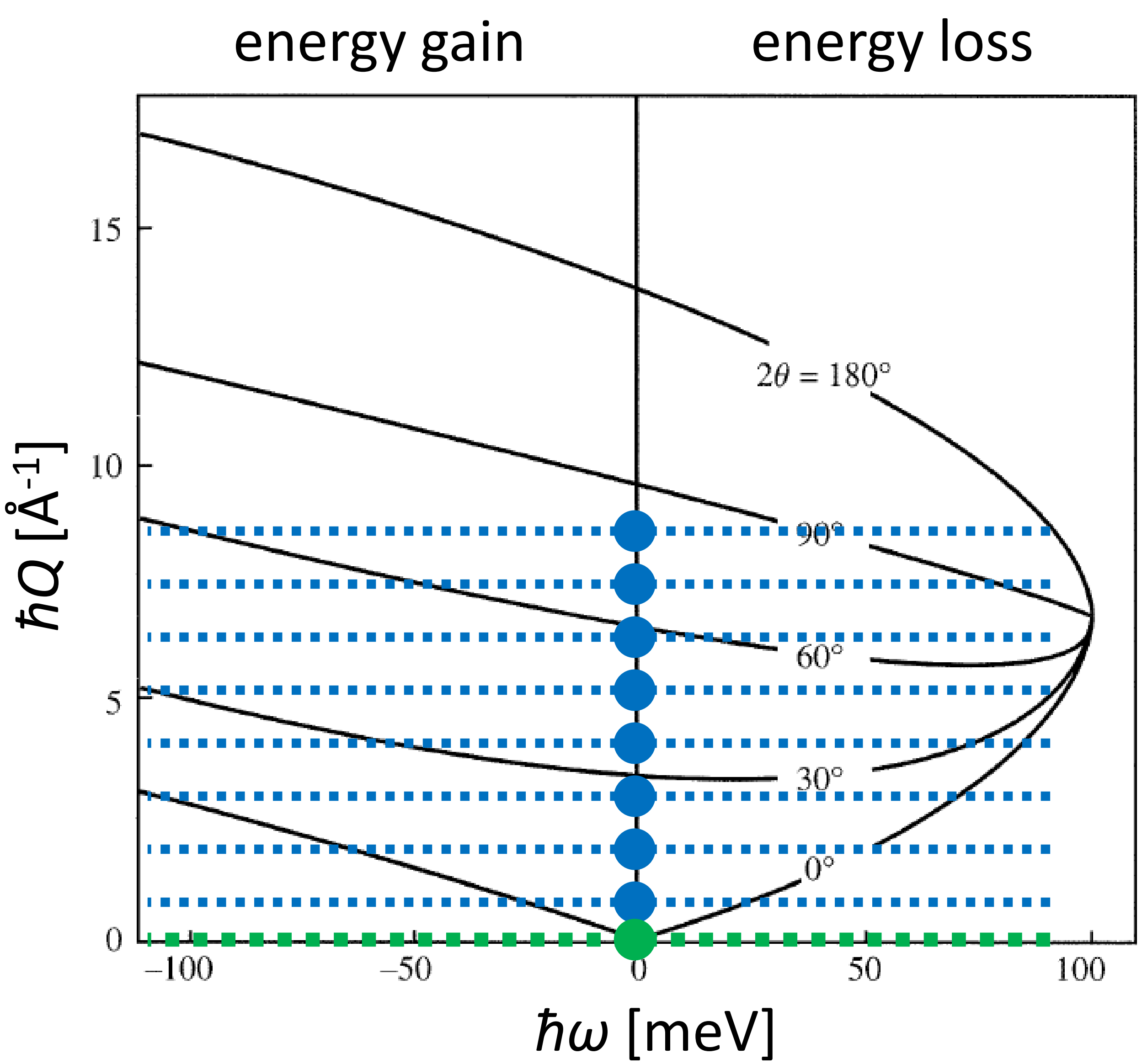
Equipped with large detector arrays to cover a wide 2θ range. Dynamic range is limited and data are recorded approximately at constant Q.



Kinematics

Kinematics of inelastically scattered x rays and visible light:

$$\vec{Q}^2 = \vec{k}_\mathrm{i}^{\,2} + \vec{k}_\mathrm{f}^{\,2} - 2|\vec{k}_\mathrm{i}||\vec{k}_\mathrm{f}|\cos(2\Theta) \qquad \hbar\omega = E_\mathrm{i} - E_\mathrm{f} = \hbar c(k_\mathrm{i} - k_\mathrm{f})$$



Energy of interest : $\hbar\omega$ = 10 meV

X rays : E_i ~ 10 keV
→ $k_\mathrm{i} = k_\mathrm{f}$
→ $\Delta E/E \sim 10^{-7}$

→ $Q = 2k_\mathrm{i}\sin(\theta)$ (~ $2\theta k_\mathrm{i}$)

Raman :
$k_\mathrm{i} = k_\mathrm{f} \sim 10^{-3}$ Å^{-1}

→ Q = 0

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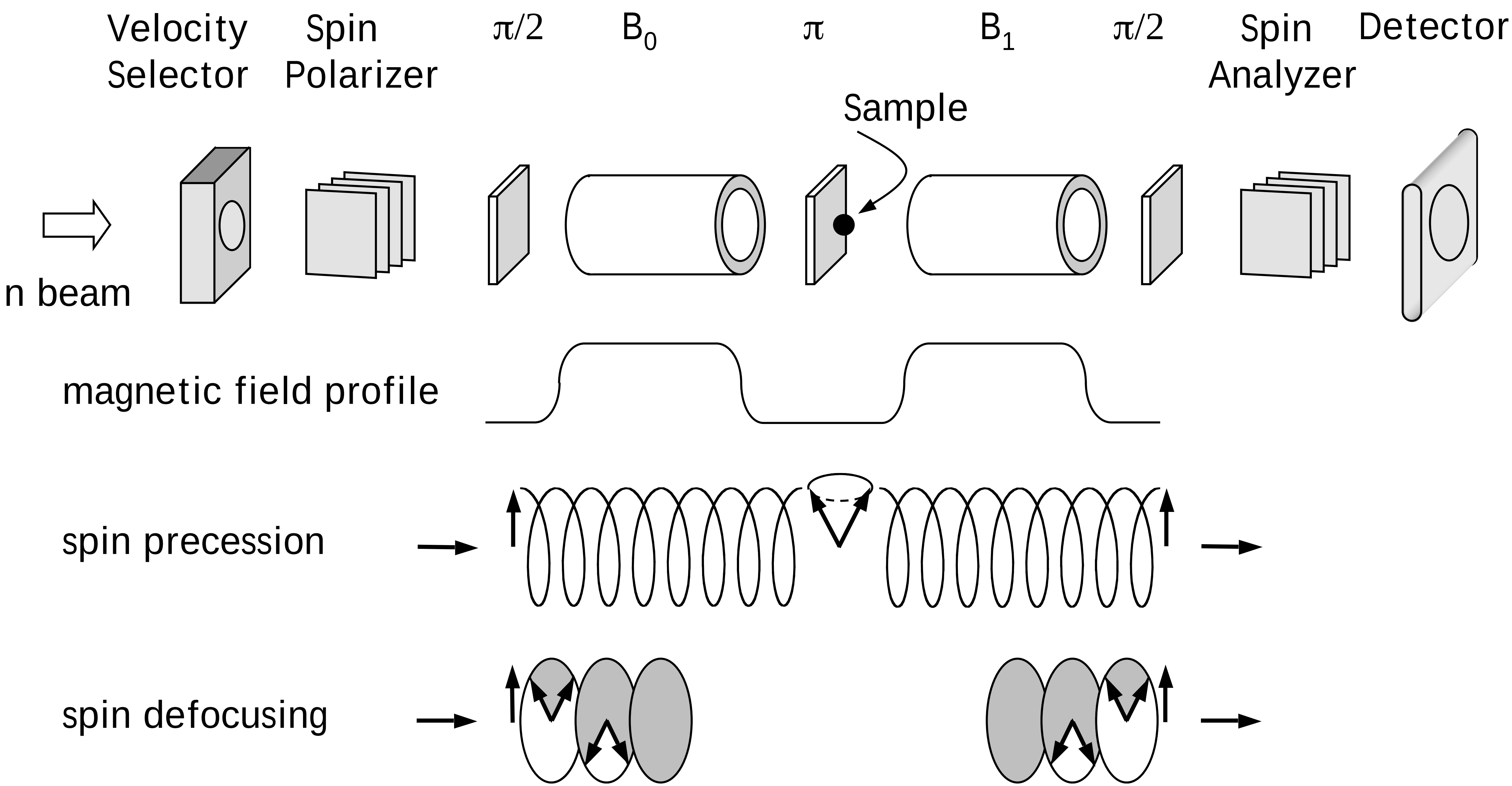
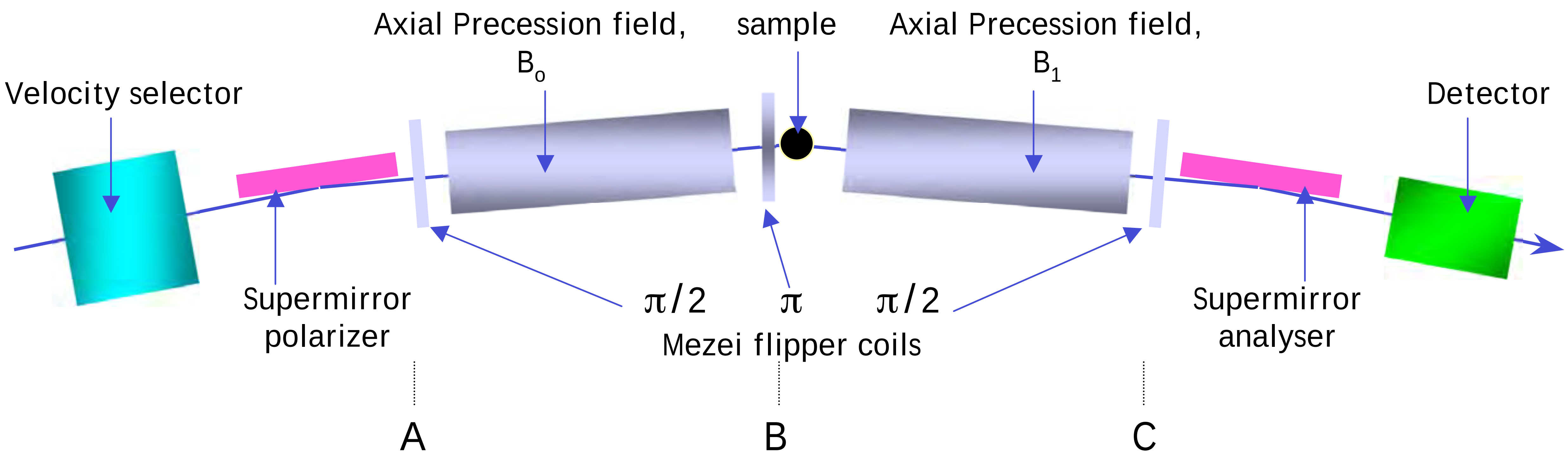
Neutron Spin Echo



NSE



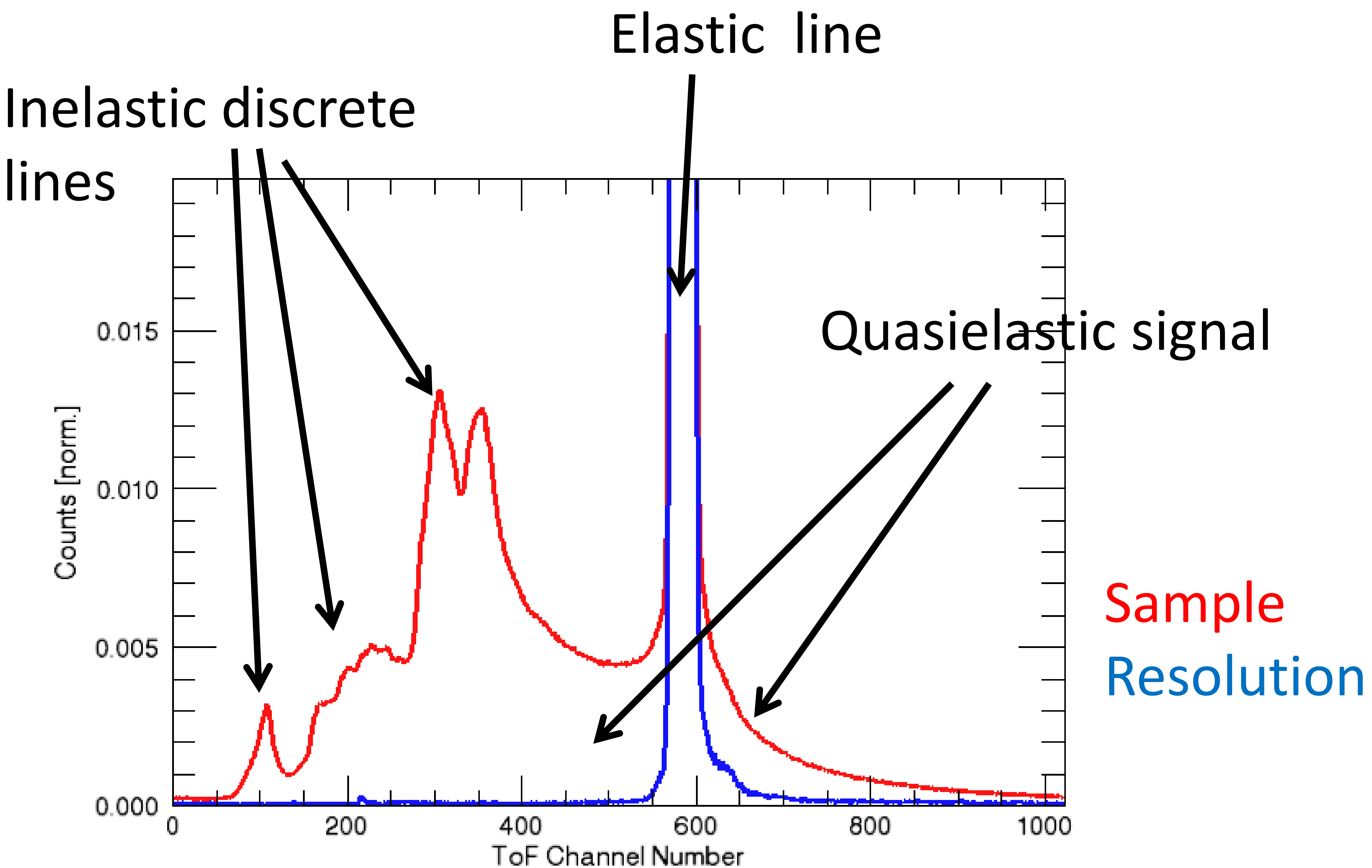
So far we have assumed that the neutrons enter and leave the spectrometer with the same distribution of velocities (i.e. any scattering by the sample is elastic)

Quasi-elastic scattering process will change the energy and therefore the velocity of the neutrons. The accumulated precession angle is now

$$\varphi_L = \gamma_n[(L_o B_o)/v_o - (L_1 B_1)/v_1]$$

Thus the accumulated precession angle φ_L can be used as a measure of the energy transfer associated with the scattering process

Elastic/inelastic scattering



Example - clathrate

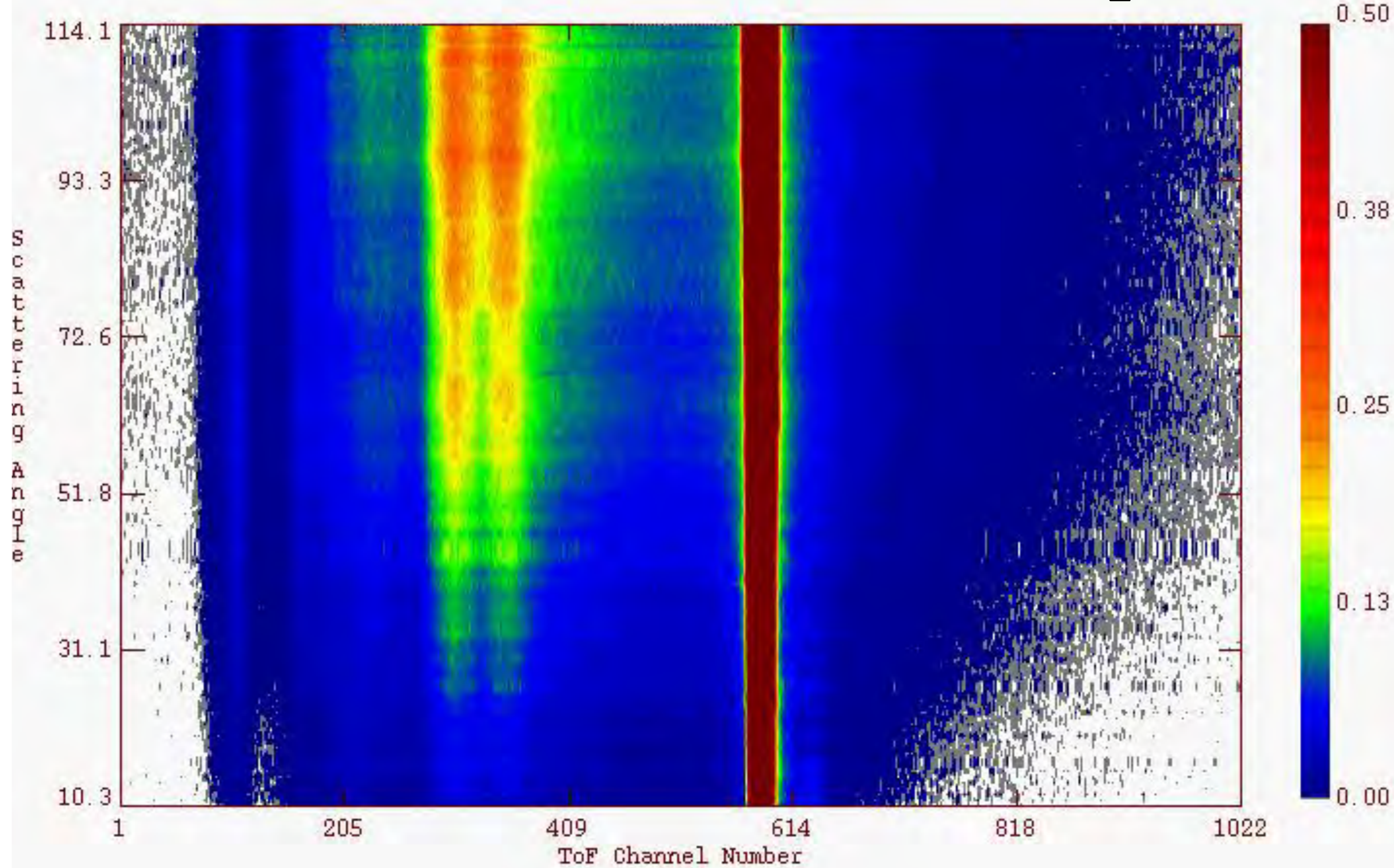
CH_4 in H_2O-clathrate:
Enormous amounts trapped in perma-frost soils and deep-sea sedimants!
Environmental risk through global warming!

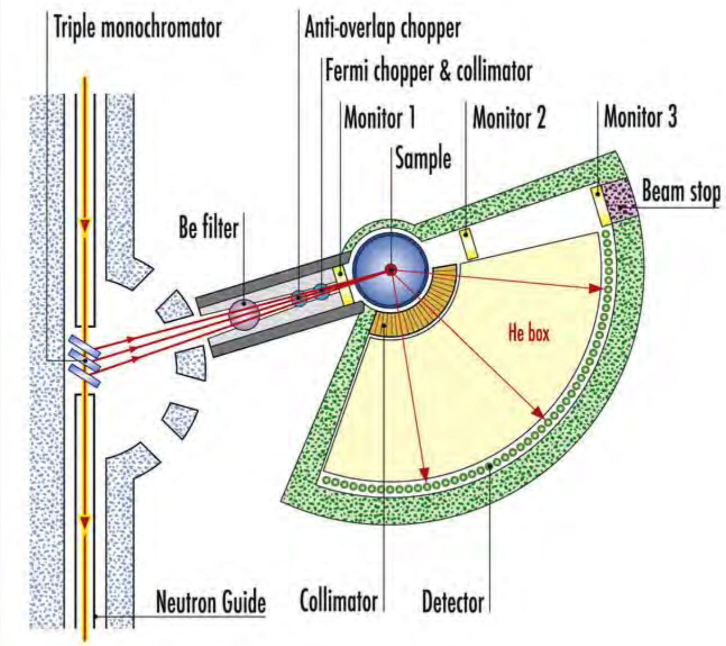
CO_2 in H_2O-clathrate :
Opportunity for dumping our combustion junk!

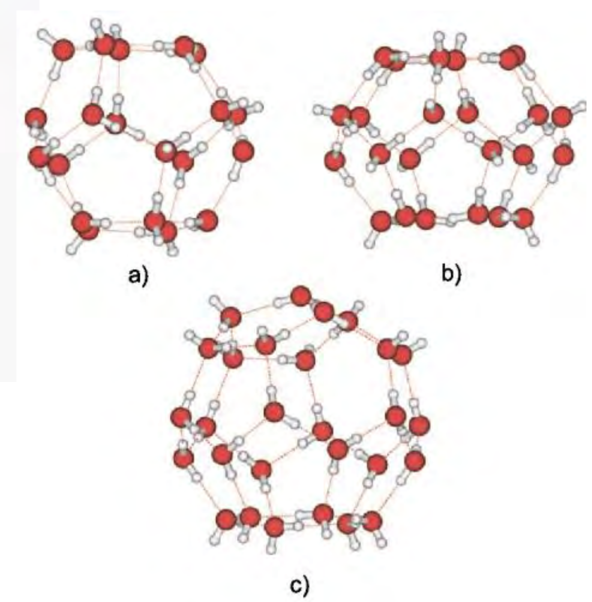
Composition – decomposition mechanism?
Stability regime and conditions?

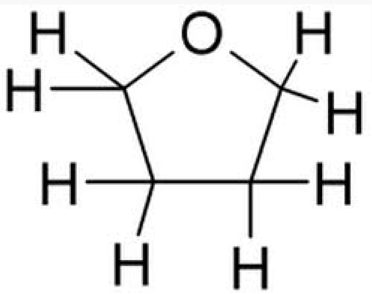
Example - clathrate

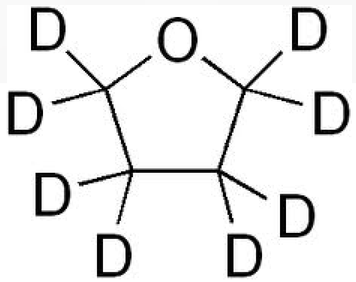
Tetrahydrofuran (THF) in H_2O hydrate-clathrate :



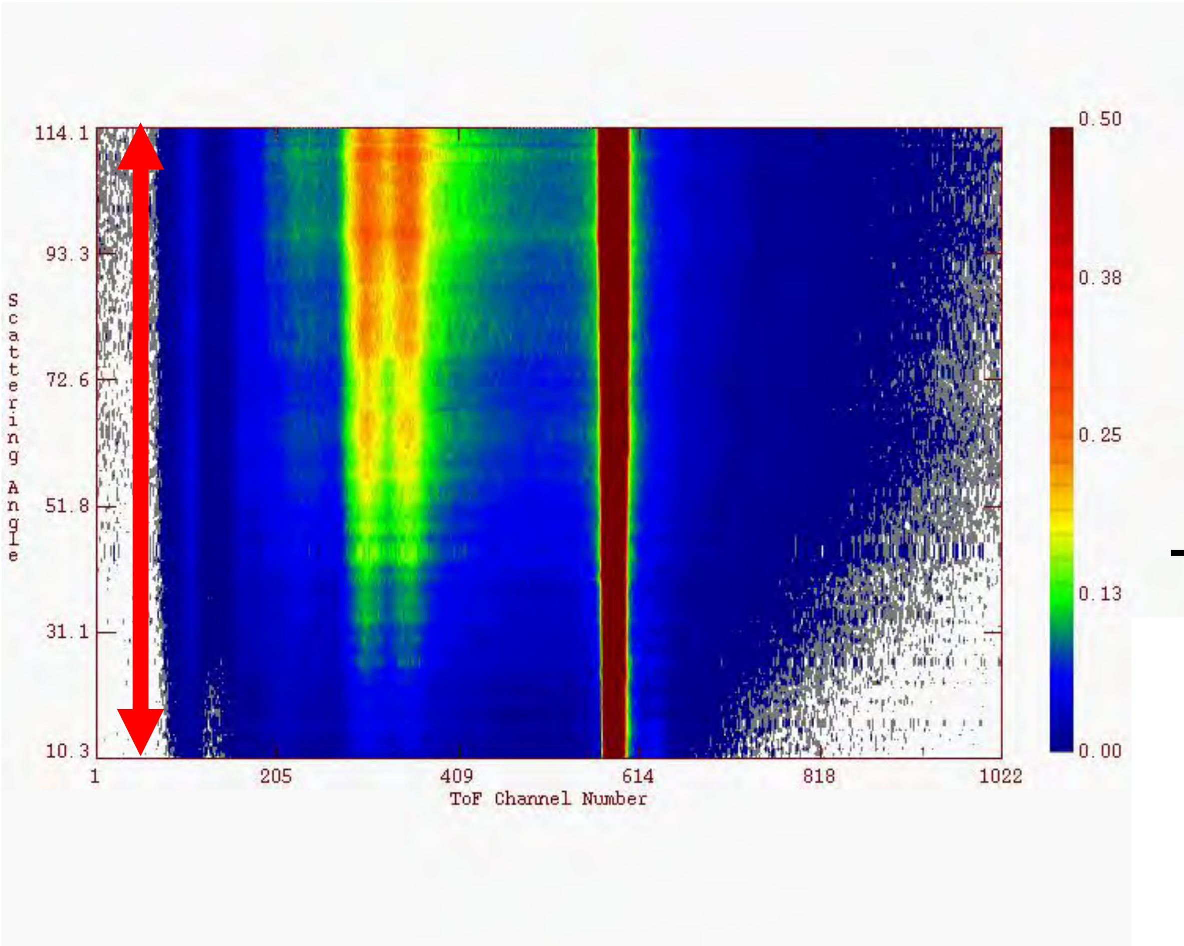








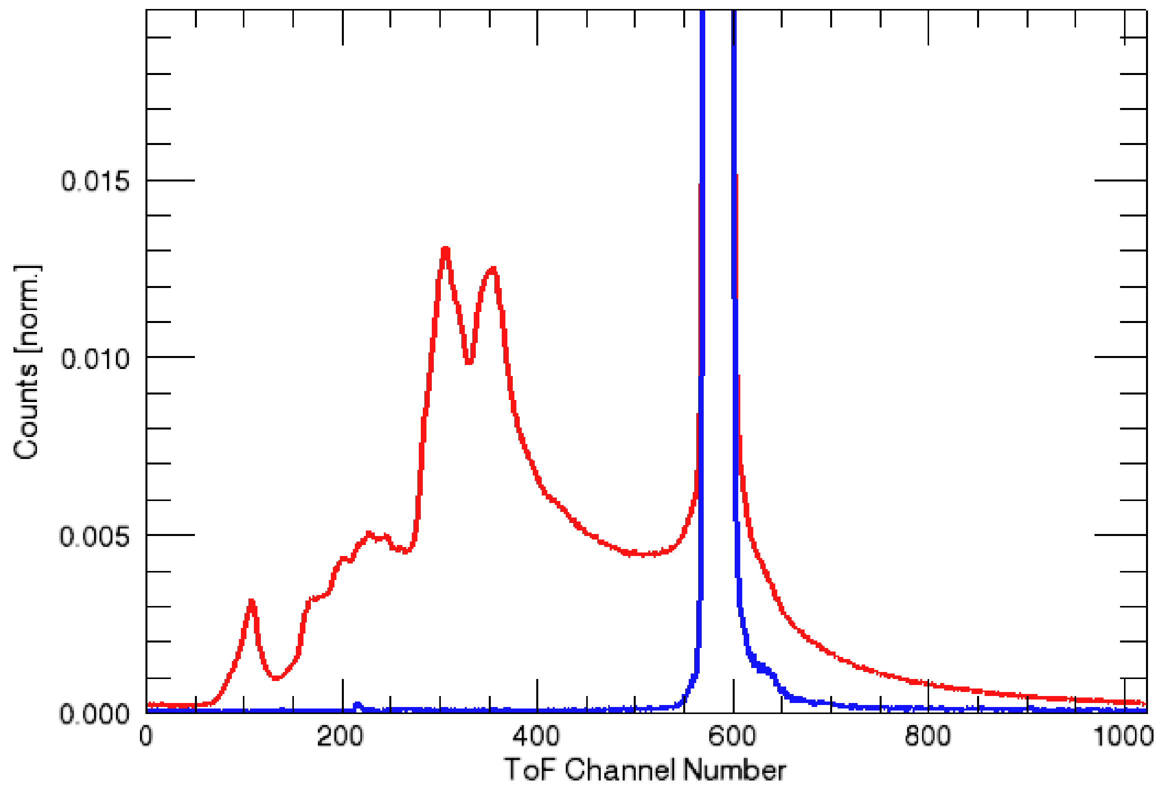
Example - clathrate



Phonon Density of States :

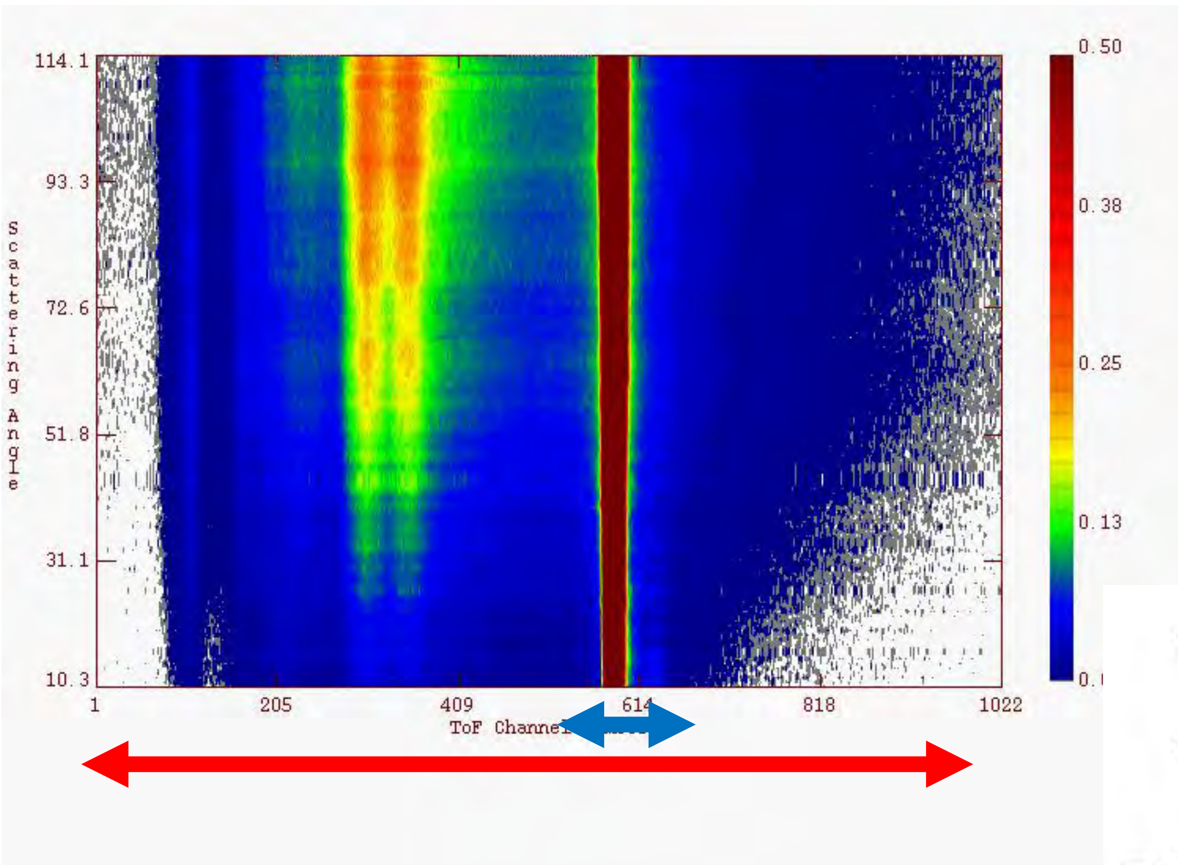
Sum up all detectors !

Text



M. Koza et al.

Example - clathrate

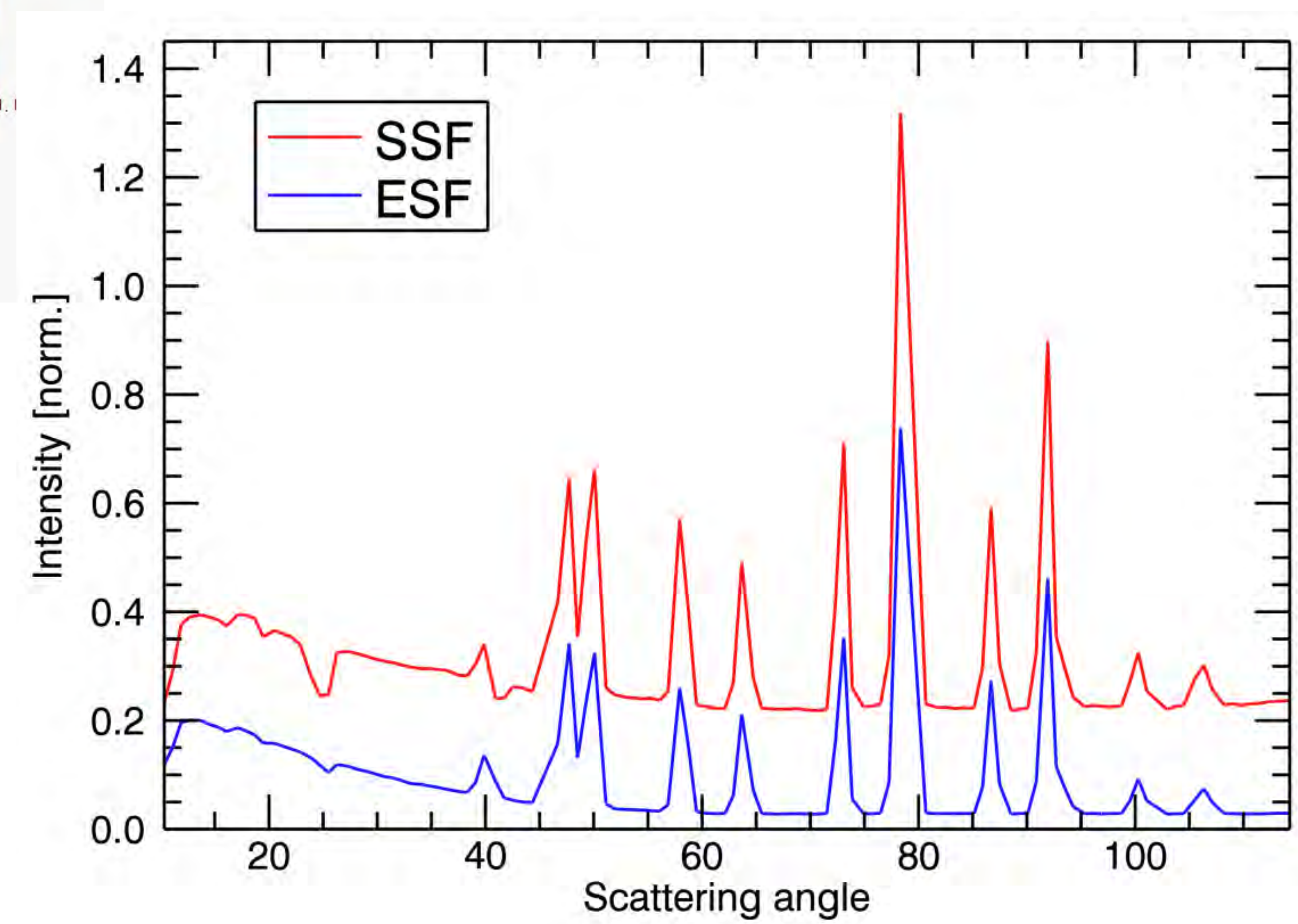


Elastic Structure Factor:

Sum up intensity in the energy channels around the elastic line only !

Static Structure Factor:

Sum up intensity in all energy channels !
DIFFRACTION!!!



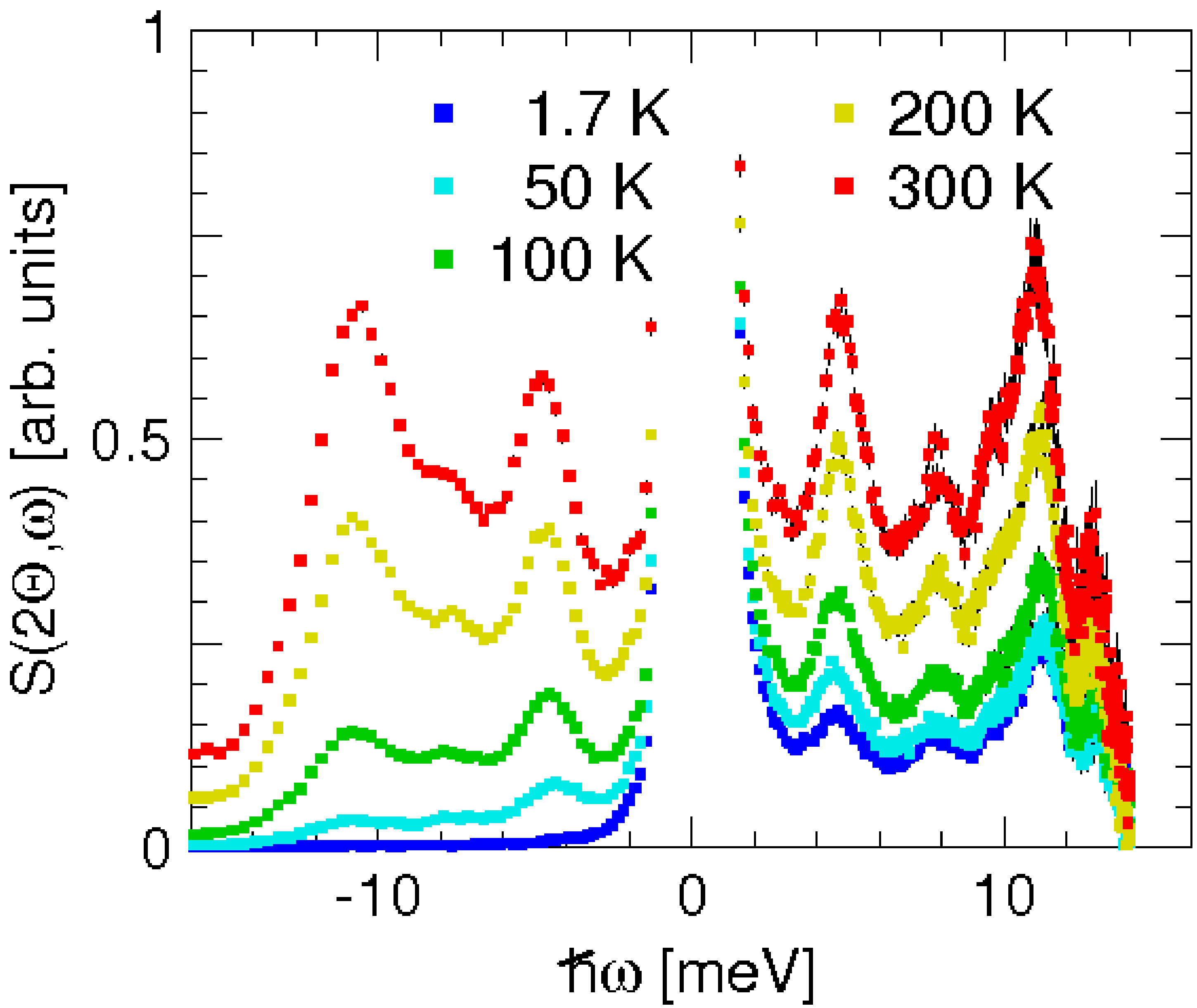
Example - detailed balance

Neutron energy gain

Anti-Stokes line

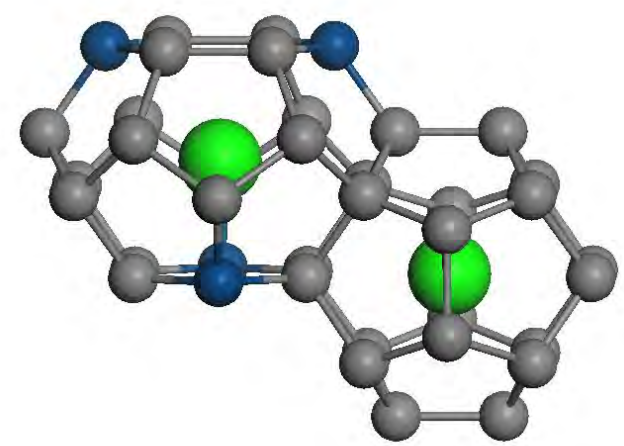
Neutron energy loss

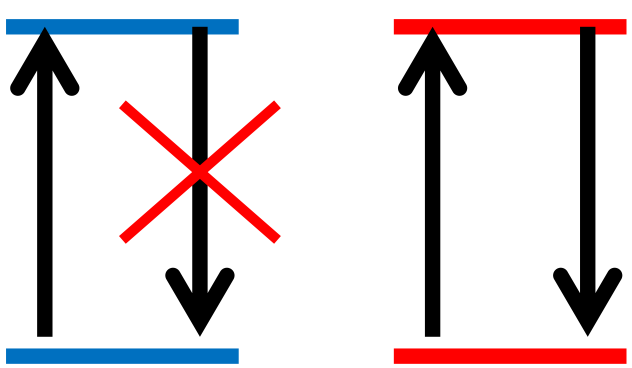
Stokes line



Clathrate :

$Ba_8Zn_xGe_{46-x}$

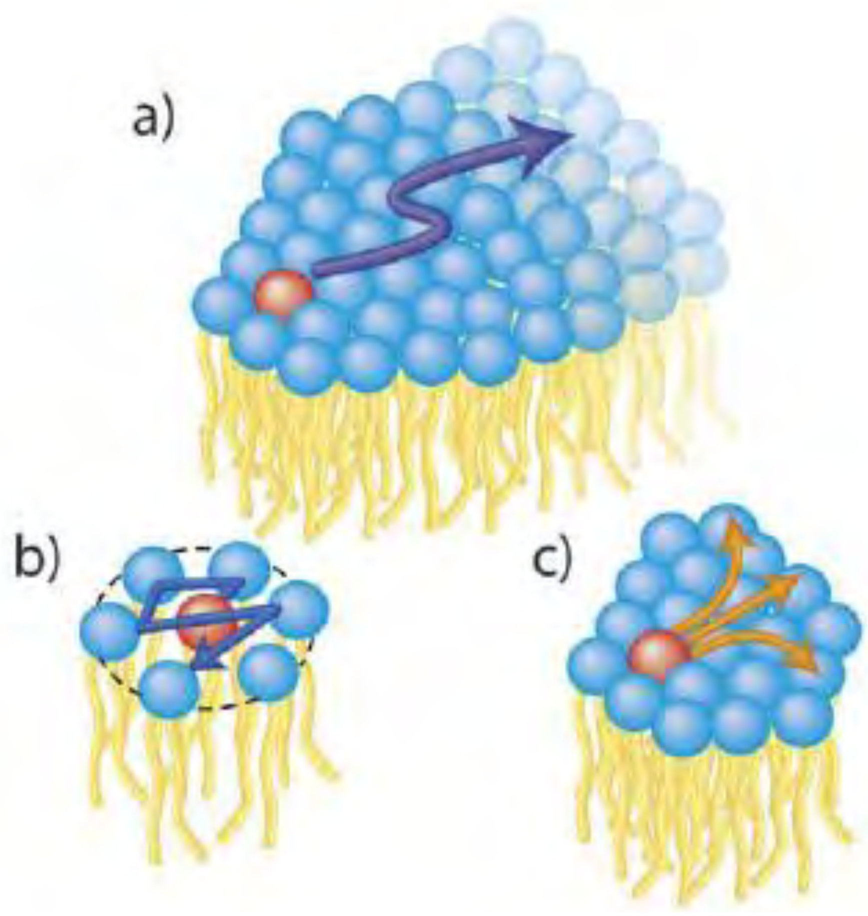




Example - ballistic motion

Short range ballistic motion in fluid lipid bilayers studied by quasi-elastic neutron scattering†

C. L. Armstrong,‡[*a] M. Trapp,‡[bc] J. Peters,[def] T. Seydel[f] and M. C. Rheinstädter[ag]

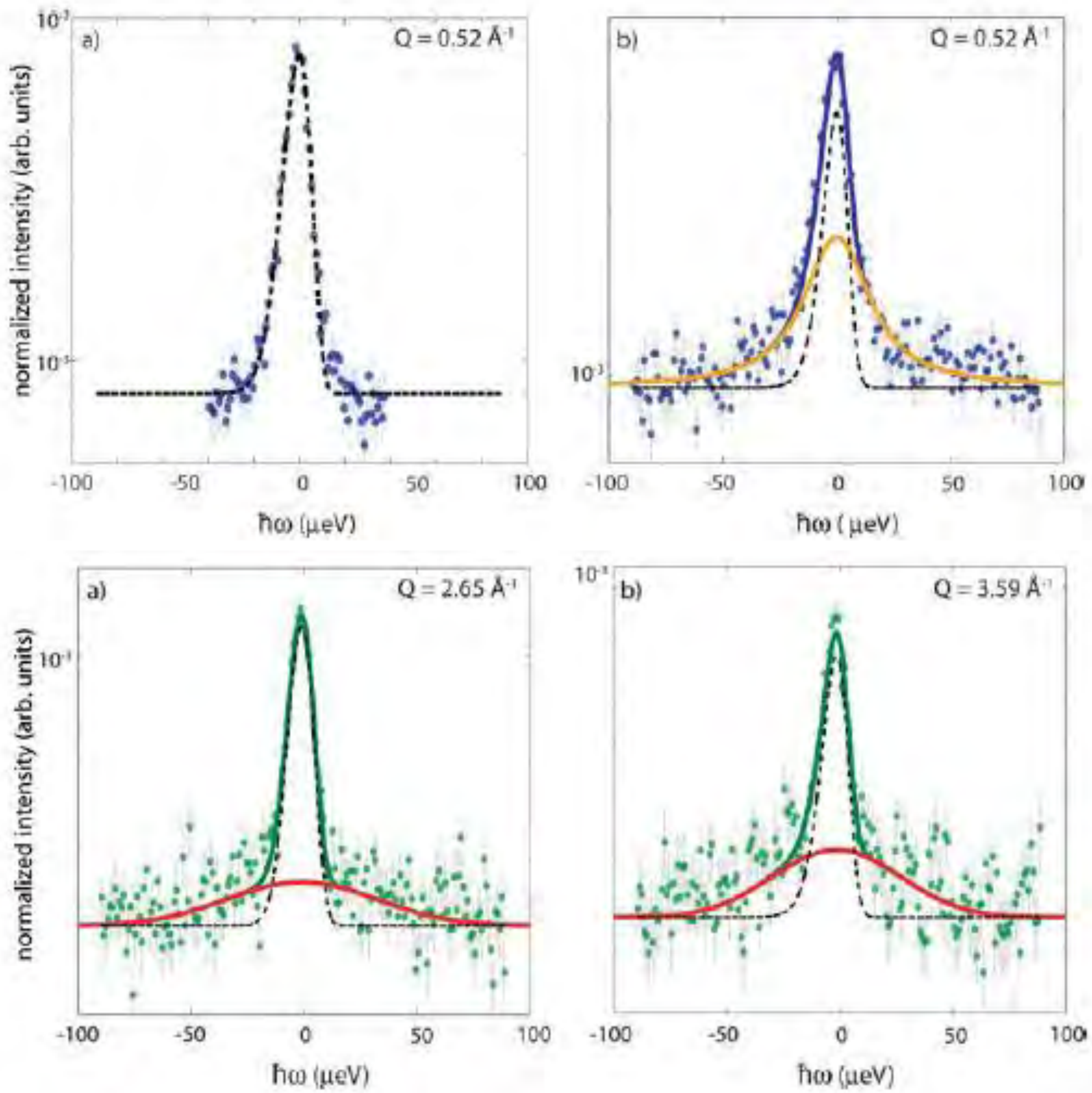


Particular interest :
Is there a change in diffusion dynamics from ballistic at short distances (time scales) to Brownian like motion at long distances (time scales) ?

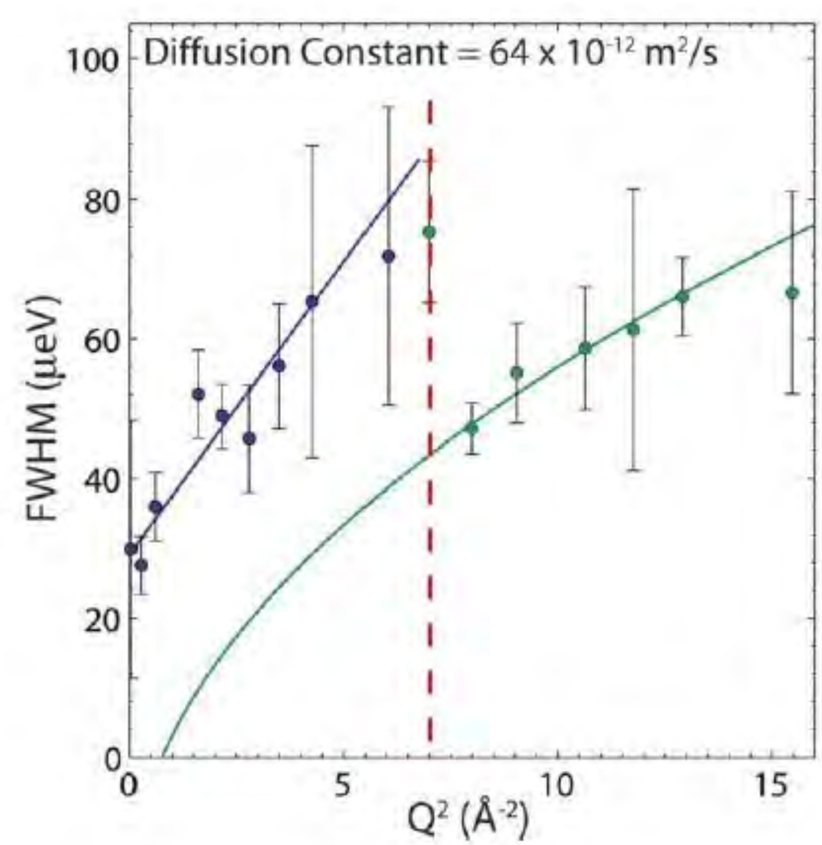
Brownian → Lorentz line

Ballistic → Gaussian line

Example - balistic motion

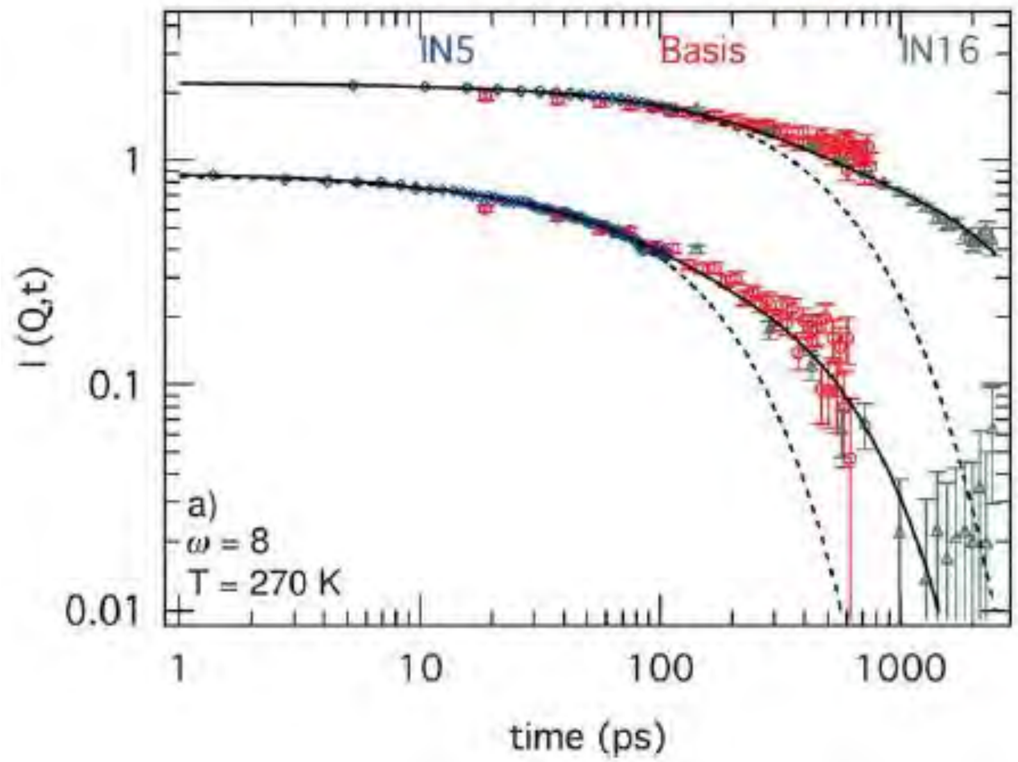


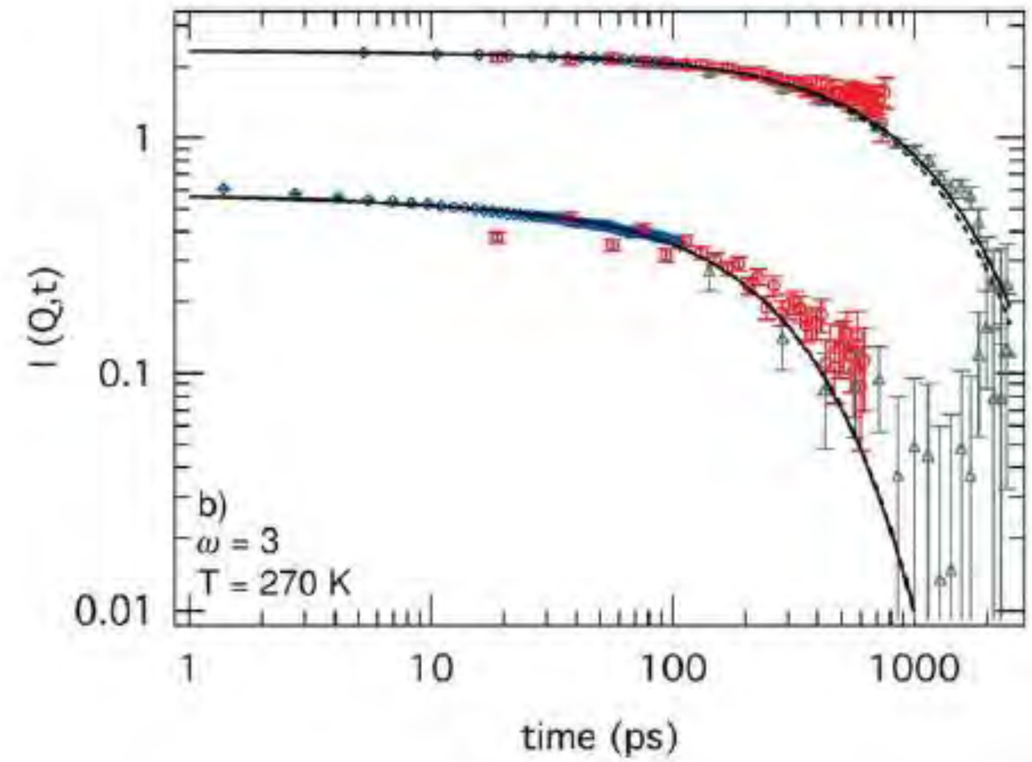
Instrument :
IN13 with cooled/heated monochromator
$\lambda = 2.2$ Å
$\Delta E = 8$ μeV
E range = -100...+100 μeV

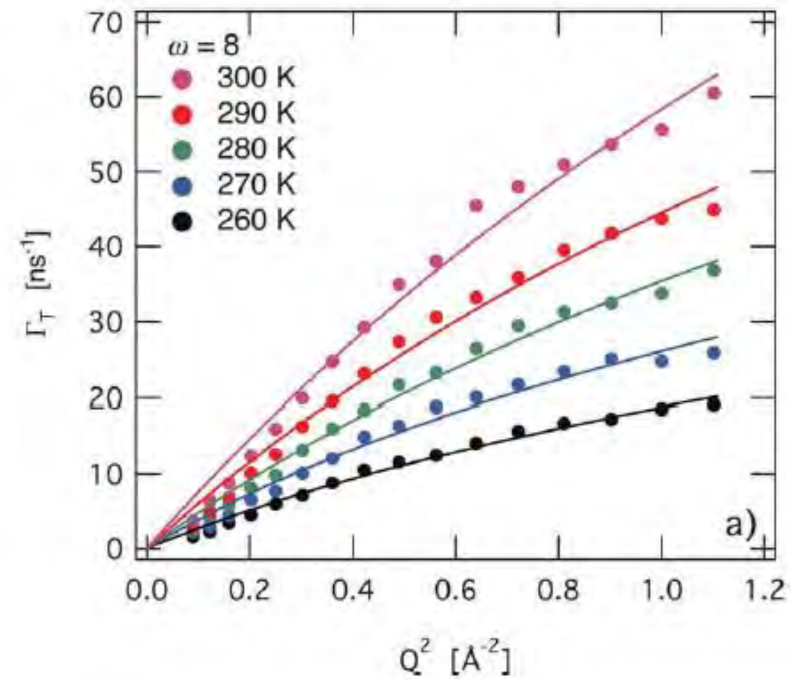


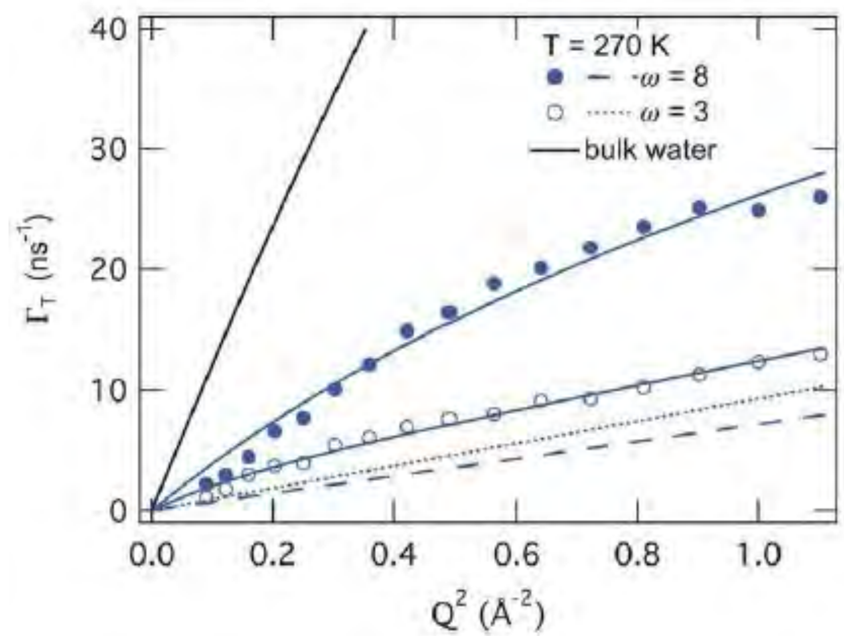
At r <2.4 Å indication of ballistic motion?

Example - confined water







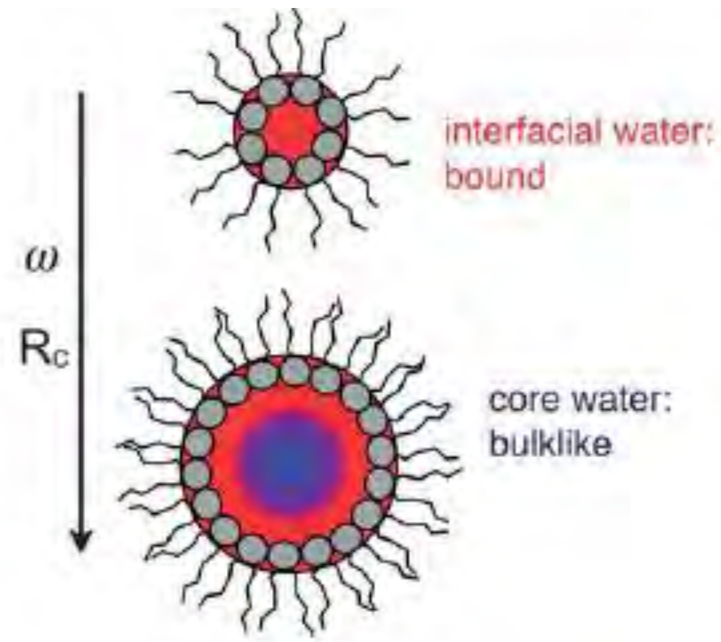


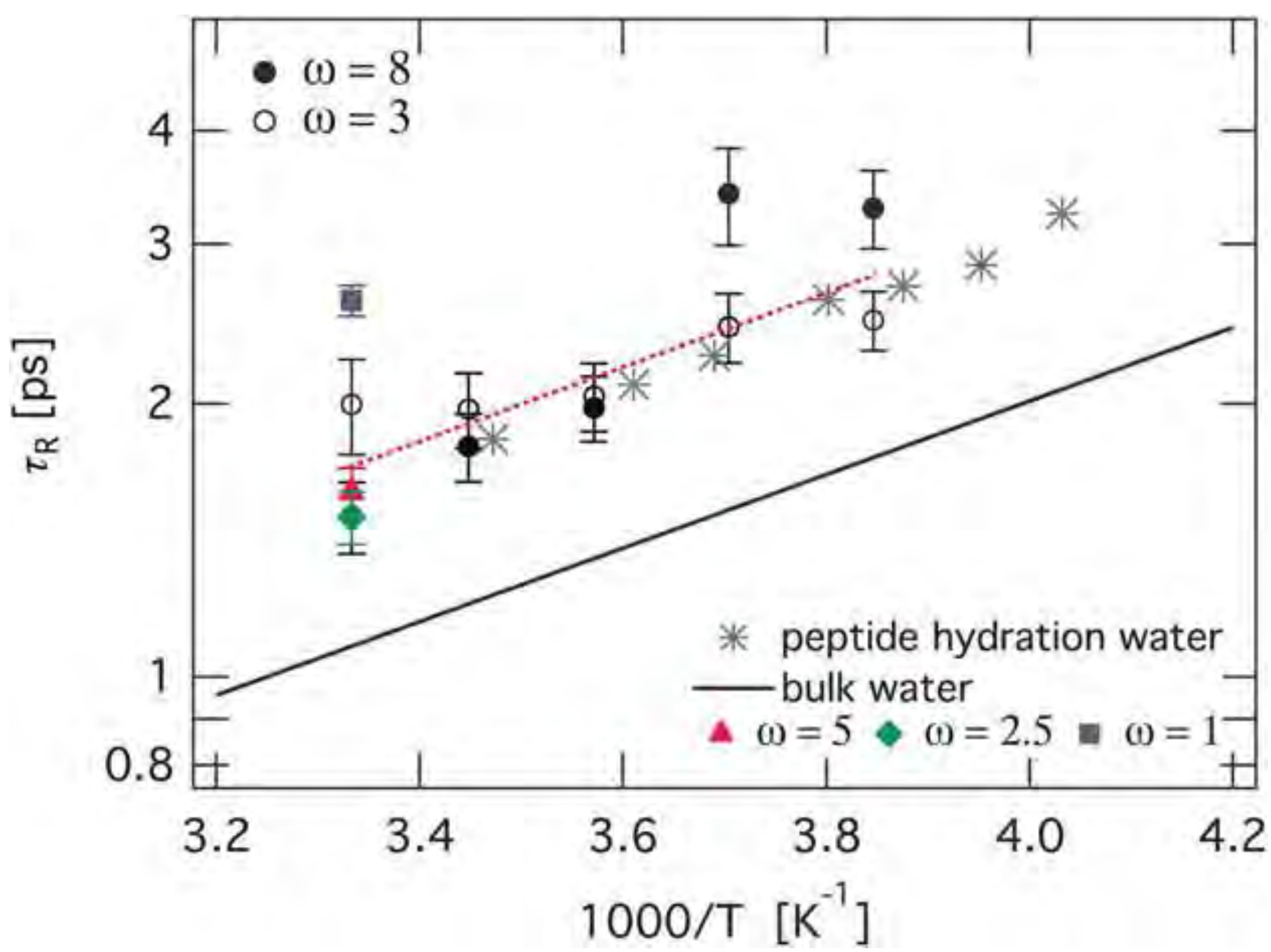
Example - confined water

Dynamics of water confined to reverse AOT micelles

Tinka Luise Spehr,[*ab] Bernhard Frick,[b] Michaela Zamponi[cd] and Bernd Stühn[a]

Size effect of micelles on diffusion properties of confined water

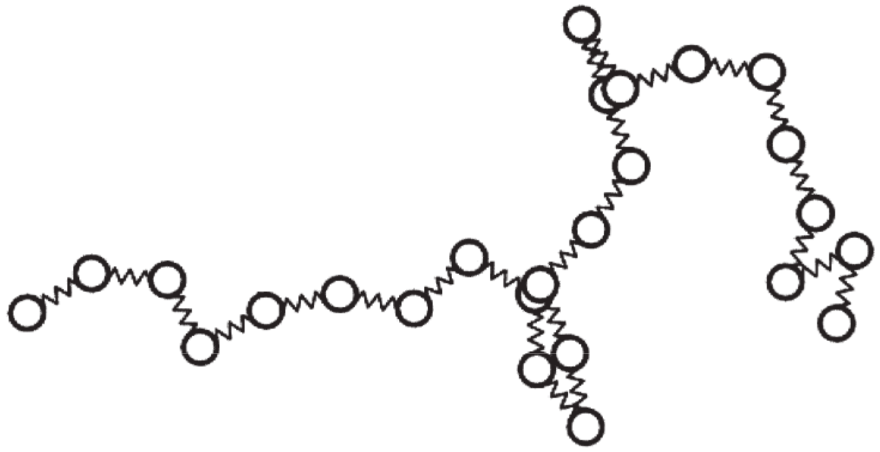




Instruments :
IN5 and IN16 at ILL
BASIS at SNS, USA

Rouse model

The current description of the Rouse dynamics is based on the bead and spring model for polymer chains.



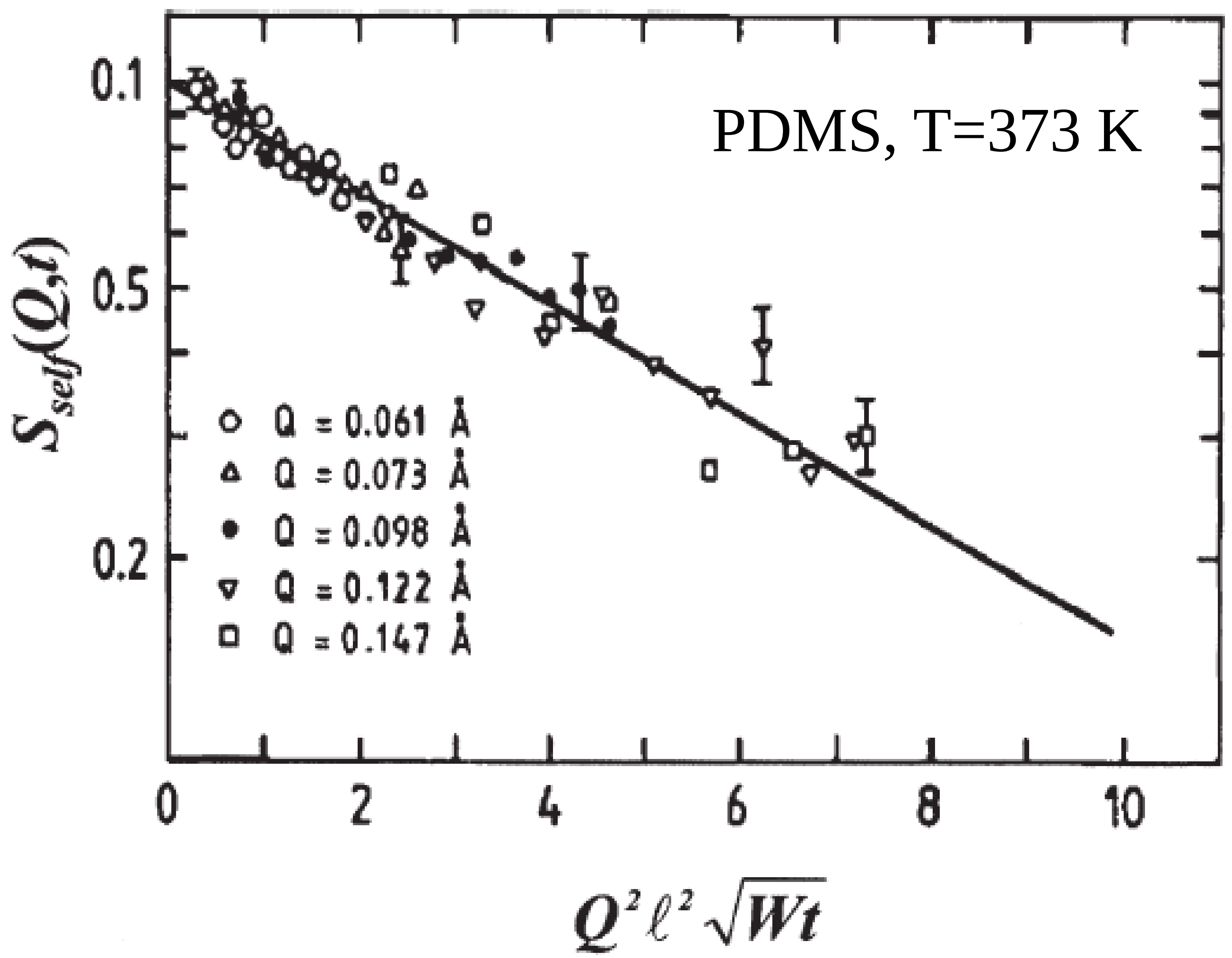
$$I_{Rouse}^{self}(Q,t) = \exp\left[-\frac{Wl^2}{3N}Q^2 t\right]\exp\left[-\sqrt{\frac{t}{9\pi}Wl^4Q^4}\right]$$

$$I_{Rouse}^{pair}(Q,t) = \frac{12}{Q^2l^2}\int_0^\infty du \exp\left\{-u-\sqrt{\Omega_R t}\,h\left[u\sqrt{\Omega_R t}\right]\right\}$$

$$h(y) = \frac{2}{\pi}\int_0^\infty dx \frac{\cos(xy)}{x^2}\left[1-\exp\left(-x^2\right)\right];\ \Omega_R = \frac{Q^4l^4}{36}W$$

NSE - rouse - self

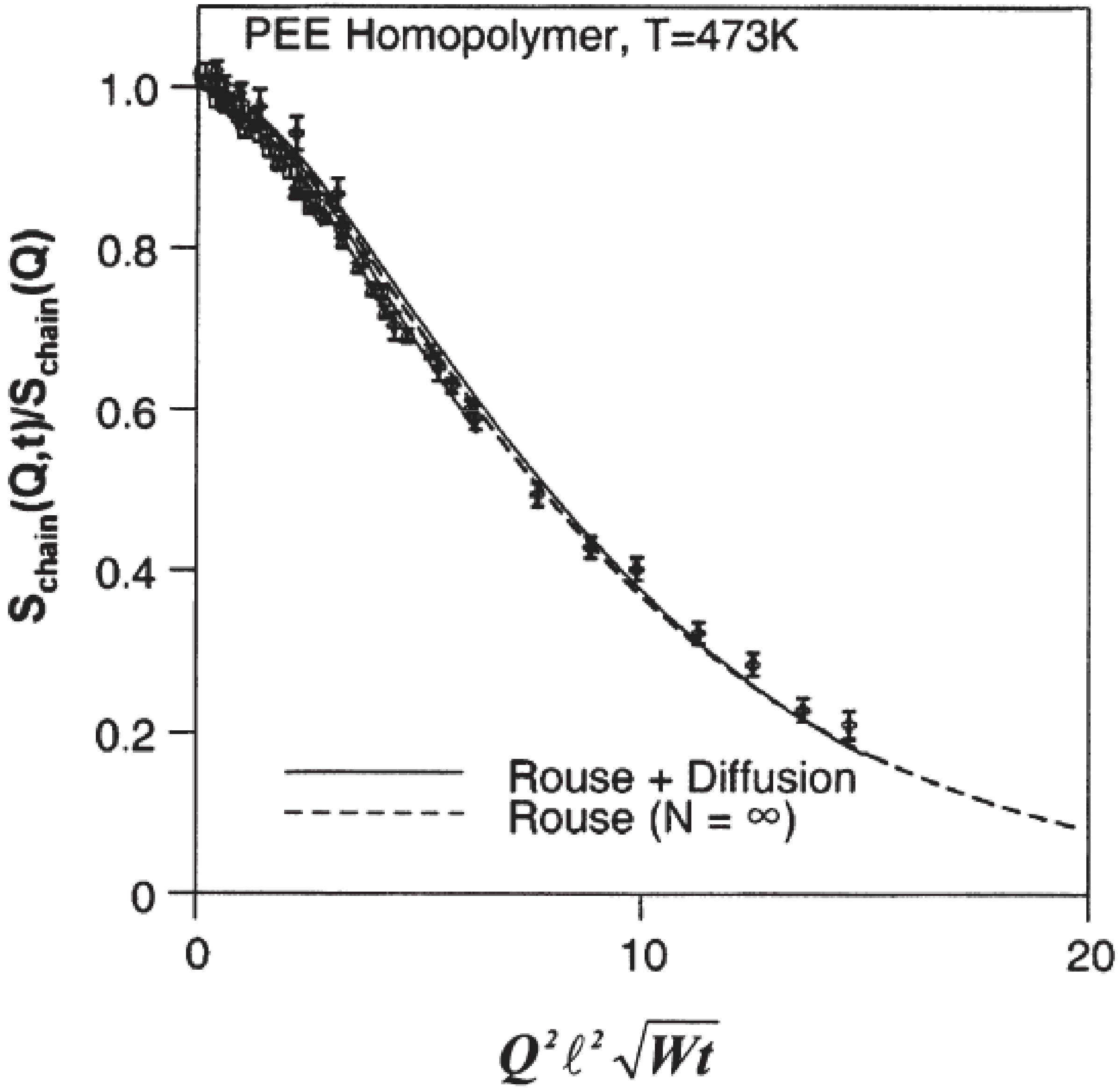
NSE results validate the theory based on Rouse dynamics:



D. Richter, et al., *Phys. Rev. Lett.*, **62**, 2140 (1989) .

NSE - rouse - pair

NSE results validate the theory based on Rouse dynamics:



H. Montes, et al., *J. Chem. Phys.*, **110**, 10188 (1999) .

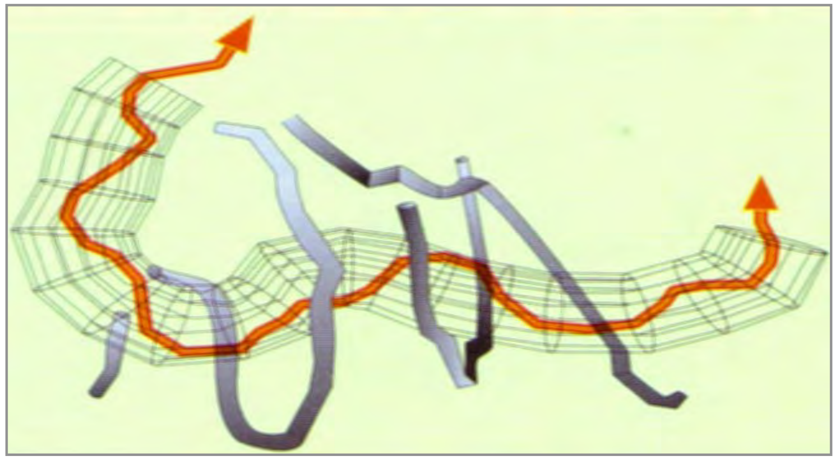
Example - Reptation

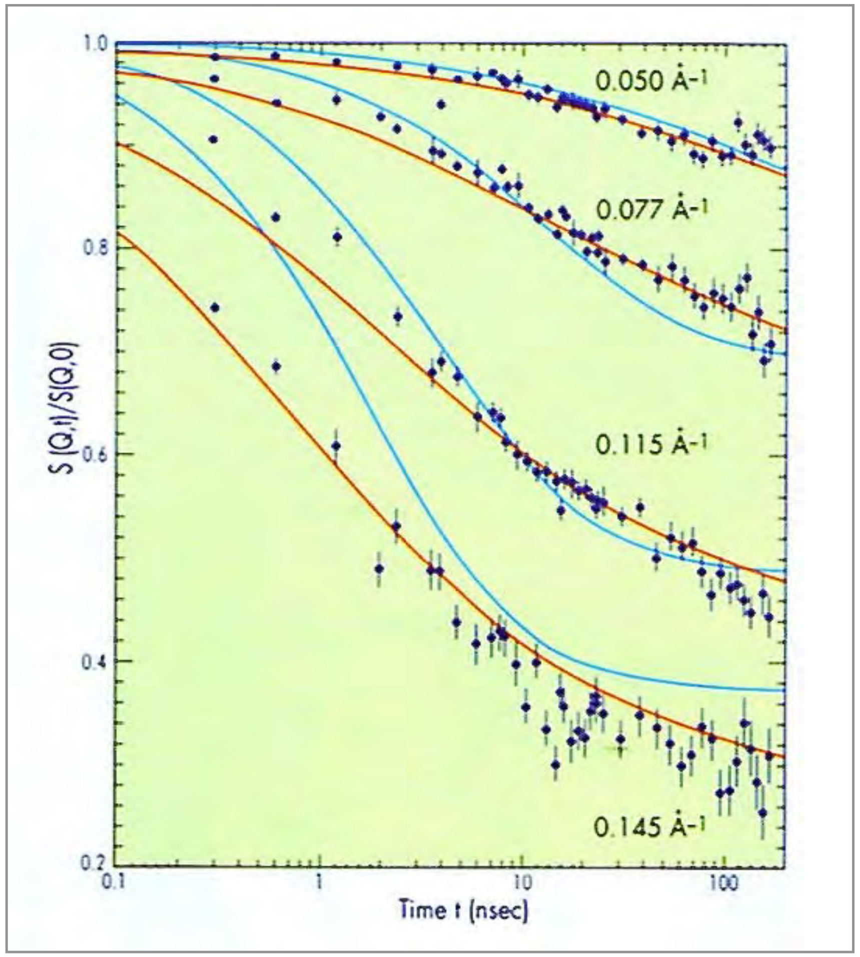
Reptation in polyethylene

The dynamics of dense polymeric systems are dominated by entanglement effects which reduce the degrees of freedom of each chain

de Gennes formulated the reptation hypothesis in which a chain is confined within a “tube” constraining lateral diffusion - although several other models have also been proposed

The measurements on IN15 are in agreement with the reptation model. Fits to the model can be made with one free parameter, the tube diameter, which is estimated to be 45Å





Schleger et al, Phys Rev Lett <u>81</u>, 124 (1998)

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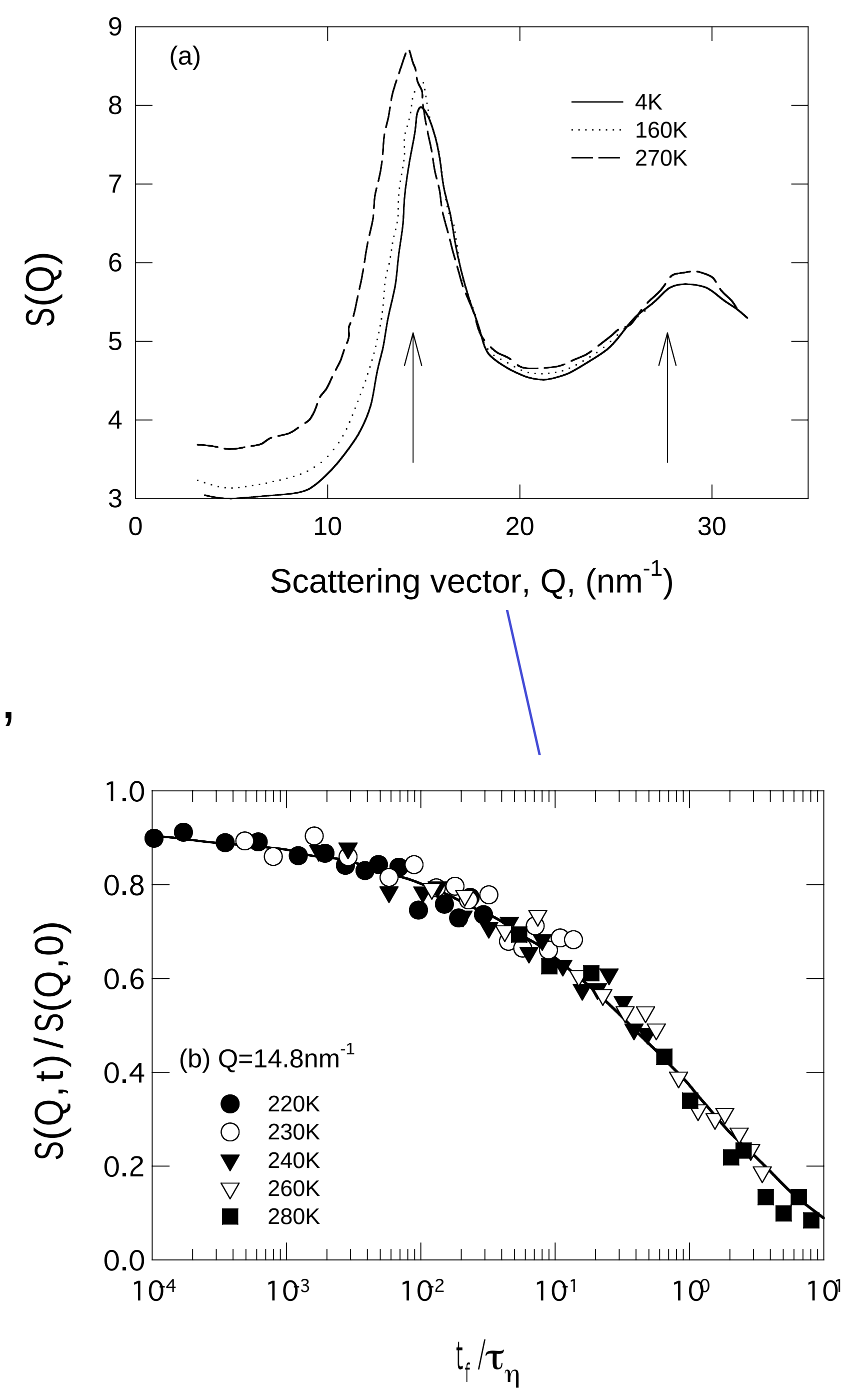
Example - glass

Glassy Dynamics in Polybutadiene

The first peak in the structure factor corresponds to *interchain* correlations (weak Van der Waals forces), the second peak is dominated by *intrachain* correlations (covalent bonding)

Each NSE spectrum was rescaled by the experimentally determined characteristic time, t_h, for bulk viscous relaxation for the corresponding temperature.

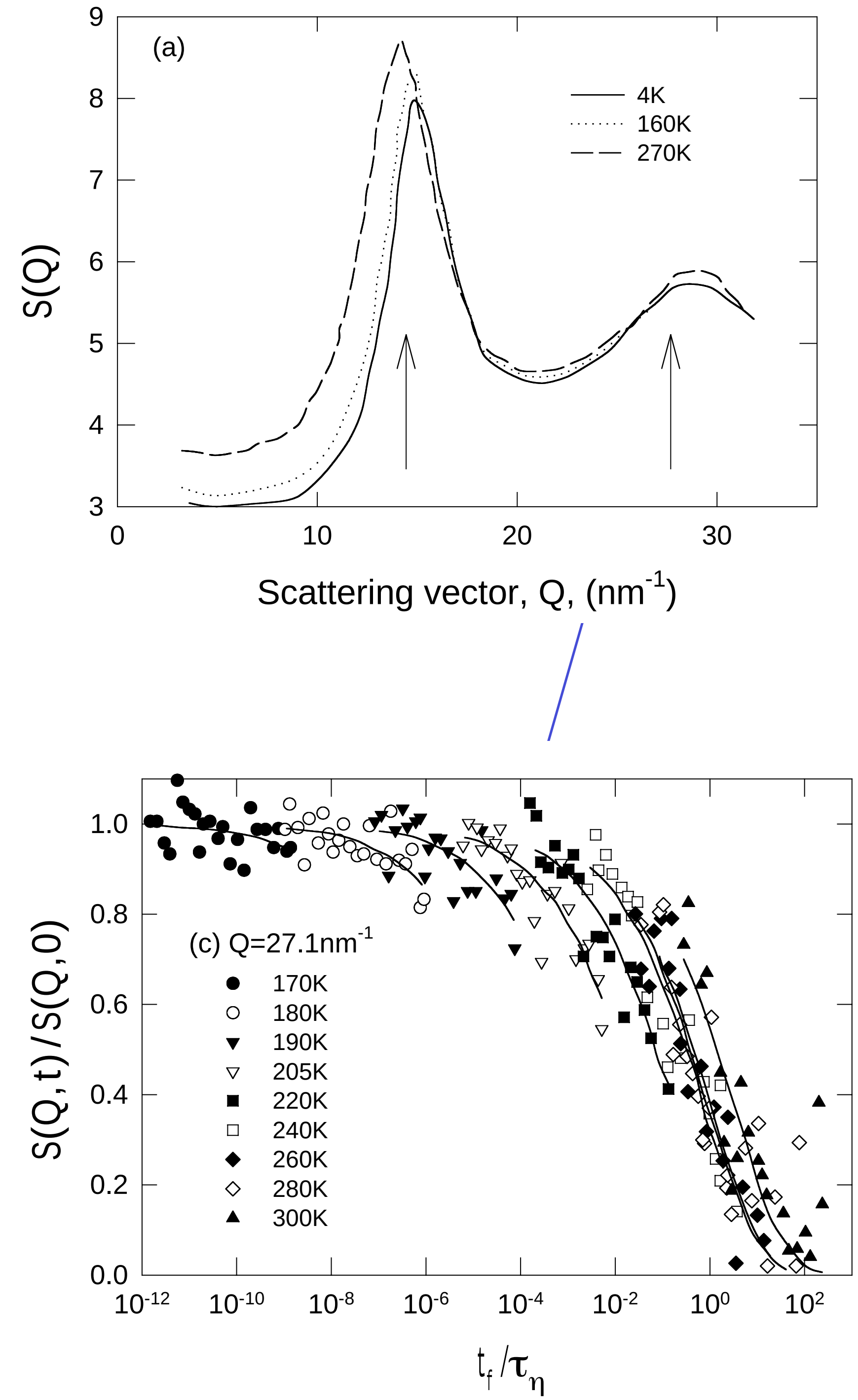
at the first peak the spectra follow a single, stretched exponential (β=0.4) universal curve, indicating that the interchain dynamics very closely follow the α-relaxational behaviour associated with macroscopic flow of polybutadiene.



Example - glass

Glassy Dynamics in Polybutadiene

Those correlation functions measured at the second peak of the structure factor do not rescale. They are characterised by a simple exponential function, with an associated temperature dependent relaxation rate which follows an Arrhenius dependence with the same activation energy as the dielectric ß-process in polybutadiene, and which is unaffected by the glass transition.



A. Arbe et al Phys. Rev. E 54, 3853 (1996)

Summary

Why is neutron scattering important?

Peculiarities of neutrons

How does it work?

What do we measure?
How do we measure?

What is it good for?

Applications